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1 **Bottom-up vs Top-down Drivers of Eruption Style: Petro-Geochemical Constraints from**  
2 **the Holocene Explosive Activity at La Soufrière de Guadeloupe**

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10 **Abstract**

11 Signals of volcanic unrest have been used successfully to provide insights into the timing,  
12 magnitude, intensity, and style of future eruptions. However, in order to provide context for  
13 the subsequent activity analysis of past eruptions is required. This provides useful  
14 information in order to understand processes of magma genesis, storage, evolution and  
15 ascent which lead to the onset of future eruptions. Here, we examine basaltic-andesitic to  
16 andesitic deposits from La Soufrière de Guadeloupe Holocene eruptions, covering a range of  
17 explosive eruption styles, ages and magnitudes. Our work is timely given unrest at this  
18 system has increased over the last 25 years, with a potential eruption capable of directly  
19 impacting up to 80,000 people in Southern Basse-Terre and potentially thousands more  
20 indirectly on a regional scale. We report on the geochemistry of pre-eruptive magmas using  
21 detailed analyses of glass (melt inclusions and groundmass glass) from four Holocene  
22 explosive eruptions: 1657 Cal. CE (Vulcanian, VEI 2), 1530 Cal. CE (sub-Plinian, VEI 3), 1010  
23 Cal. CE (Plinian, VEI 4), and 5680 Cal. BCE (Plinian, VEI 4). Major element concentrations vs  
24 SiO<sub>2</sub> in whole rock (WR), groundmass glass (GM) and melt inclusions (MI) show a strong  
25 linear trend. MIs reveal a relatively homogenous melt composition from the first to the  
26 most recent eruptions, ranging from 63.6 – 78.7 wt% SiO<sub>2</sub>. Volatiles, including H<sub>2</sub>O (2.3 - 4.4  
27 wt%), CO<sub>2</sub> (35 - 866 ppm) and sulphur (30 - 202 ppm), are also consistent across the various  
28 eruptions. The major element and volatile compositional homogeneity across the eruptions  
29 indicates that composition and volatiles do not have a direct control on eruption explosivity  
30 at this system. Instead, we find differences in ascent rate, groundmass glass viscosity and  
31 microlite volume percentage indicating that explosive eruptive style at La Soufrière is  
32 controlled by a combination of ascent rate and top-down controls affecting rock strength,  
33 stress distribution and the development of fluid overpressure. Rapid ascent in the absence  
34 of top-down controls (processes with a cause external to the magma but affecting the  
35 plumbing system) will result in explosive eruptions driven from the bottom-up (internal to  
36 magma dynamic response with varying pressure and temperature, e.g., 1010 Cal. CE in the  
37 case of very rapid ascent or 1657 Cal. CE in the case of rapid ascent). However, we also  
38 highlight the importance of top-down controls such as conduit sealing which can promote

39 the onset of explosive eruptions, even in the case of slow magma ascent (e.g., 5680 Cal.  
40 BCE). External effects (including ingress of water and rapid edifice unloading) can also favour  
41 explosive eruptions with flank collapses involved in some scenarios (e.g., 1530 Cal. CE). The  
42 multiple controls on explosive eruption style make this system more hazardous and complex  
43 to model and monitor. In order to improve early-warning system efficiency, forecast  
44 models, eruption scenario crisis response and long-term risk reduction planning, we stress  
45 that internal processes such as fracture and host-rock sealing (fluid pore pressure) as well as  
46 external processes such as water moving into the system and the mechanical stability of the  
47 edifice should be monitored and modelled closely.

48 **Keywords:** Eruption style, melt inclusions, ascent rate, mixing and mingling, La Soufrière de  
49 Guadeloupe

## 50 **1 Introduction**

51 Eruption style dictates many of the hazards presented by a volcano, however, volcanoes are  
52 not limited to one particular eruption style. Changes in eruption style can occur over short  
53 periods e.g., multiple transitions within a single eruptive episode (e.g., Cioni, et al., 2008;  
54 Boudon et al., 2008) and long periods e.g., different eruption styles separated by a repose  
55 period (e.g., Endo et al., 1981; Ratdomopurbo et al., 2013; Costa et al., 2013). In particular,  
56 explosive eruptions are hazardous and can range in style at one system from small  
57 Vulcanian activity to large sub-Plinian and Plinian activity (e.g., Soufrière Hills Volcano,  
58 Montserrat, (Komorowski et al., 2010; Wadge et al., 2014); La Soufrière de Guadeloupe  
59 (Komorowski et al., 2005; Legendre, 2012); Etna, Italy (Coltelli et al., 2005; Moretti et al.,  
60 2018; Cappello et al., 2019); Rabaul, Papua-New-Guinea (Bernard and Bouvet de  
61 Maisonneuve, 2020)).

62 Current volcano monitoring, such as geochemical and geophysical methods, can provide  
63 indications of volcano activity and the onset of unrest (e.g., Gottsman et al., 2019; Pritchard  
64 et al., 2019). However, in order to provide a more complete picture, understanding the  
65 controls on eruptive style, and particularly the controls on transitions between styles is  
66 required to develop realistic eruption scenarios.

67 There are multiple ways to generate a magmatic, explosive eruption: (1) heating of the  
68 system causing magma convection and vesiculation (Sparks, et al 1977; Melnik and Sparks,  
69 1999) and/or the mobilization of crystal-rich magmas (Burgisser and Bergantz, 2011;  
70 Andrews and Manga et al., 2014); (2) addition of magma volume into the system which  
71 increases the overpressure on the confining walls (Ruprecht and Bachmann, 2010; Bouvet  
72 de Maisonneuve et al., 2013; Degruyter, et al 2016); (3) effective degassing of volatile rich  
73 magma and volatile fluxing which increases buoyancy and overpressure during ascent  
74 (Roggensack et al., 1997; Scandone et al., 2007; Helo et al., 2011; Sides et al., 2014; Costa et  
75 al., 2013; Parmigiani et al., 2016; Edmonds and Wallace, 2017); (4) rapid magma ascent rate  
76 preventing efficient gas–melt segregation (e.g., Gonnermann and Manga, 2007; Goepfert  
77 and Gardner, 2010; Martel, 2012; Lloyd et al., 2014; Barth et al., 2019); (5) syn-eruptive

78 microlite growth resulting in an increase in magma viscosity (e.g., Houghton & Gonnermann,  
79 2008; Moitra et al., 2018; Arzilli et al., 2019; Hlinka, 2020; Hlinka et al., 2021), (6) increased  
80 mechanical connectivity by shallow level volcanic seismic rupturing (Roman and Cashman,  
81 2018). These processes are likely to occur combined in order to produce magma  
82 fragmentation and explosive eruptions (Gonnermann and Manga, 2007; Cassidy et al., 2018;  
83 Hlinka, 2020; Hlinka et al., 2021).

84 At subduction zones transition in eruption style is common, with transitions observed within  
85 the same eruption (e.g., effusive to explosive) and across several eruptions with an  
86 observed repose period (Cassidy et al., 2018). The range of eruption styles observed at such  
87 systems makes them particularly hazardous and defining the processes and controls on each  
88 eruption style is particularly important. In recent years the interplay of bottom-up (internal  
89 to magma dynamic response with varying pressure and temperature) vs top-down  
90 (processes with a cause external to the magma but affecting the plumbing system) has been  
91 shown to play an important role in triggering transitions in eruption style (e.g., Sparks, 1997;  
92 Cassidy et al., 2018; Bernard and Bouvet de Maisonneuve, 2020).

93 Bottom-up processes drive an eruption through physico-chemical changes which are  
94 intrinsic in the magma system (e.g., crystallisation, degassing and chemical differentiation,  
95 mixing of other magmas) and control magma rheology. These processes affect magma  
96 properties, and therefore affect magma migration in the crust and ascent and  
97 decompression rates, key parameters in determining eruption style, particularly the  
98 explosive one via the onset of fragmentation (Dingwell, 1996; Alidibirov and Dingwell, 1996;  
99 Gonnermann, 2015 and refs. therein). For a given felsic magma composition the total  
100 amount of volatiles in the system and the composition of the melt itself, are critical  
101 parameters in determining fragmentation thresholds and explosivity (e.g., Papale, 1999; Di  
102 Genova et al., 2017).

103 Top-down processes may involve non-magmatic causes and sources, related to water  
104 circulation and gravity or be the result of magma evolution in conduits. Rapid and efficient  
105 infiltration of external waters can lead to phreato-magmatism (Zimanowski et al., 2015);  
106 over a longer-term, water circulation promotes rock alteration and secondary  
107 mineral precipitation in the presence of well-developed hydrothermal systems (e.g., Barberi  
108 et al., 1992; Roman et al., 2019; Stix & de Moor, 2018). Top-down processes may greatly  
109 modify the edifice stability and rock, strength, and/or promote fluid overpressure. In all  
110 cases they alter the stress distribution within the volcanic edifice, particularly in the upper  
111 portion at shallow depths.

112 Gravity-induced instabilities can determine sudden changes to the edifice e.g., flank collapse  
113 or dome collapse resulting in a downward-propagating decompression wave which triggers  
114 magma fragmentation (e.g., Voight et al., 1981; Sparks et al., 2002; Alidibirov and Dingwell,  
115 1996; Komorowski et al., 2013). On longer timescales, volcano spreading was recognized to  
116 decrease thresholds for the occurrence of highly-explosive Plinian eruptions (Borgia et al.,  
117 2005).

118 Top-down controls resulting from magma evolution in conduits involve sealing of the upper  
119 portions of a plumbing system, which results from rapid thermal exchange due to a high  
120 surface/volume ratio when magma supply is low or nil and degassing-induced crystallization  
121 is high (De Natale et al., 2004; Quarenì et al., 2007). Concomitant but external factors  
122 contributing to conduit sealing are plugging due to collapse (particularly at calderas) and the  
123 rock overburden pressure, which the tectonic regional stress can contribute to either  
124 positively (compressive) or negatively (tensile) (Quarenì and Mulargia, 1993). Changes that  
125 occur in the uppermost plumbing system affect late-stage decompression path of magma  
126 considerably and control eruption style from the top-down (Thomas and Neuberg 2014,  
127 Cassidy et al., 2018). An open conduit, or one which is not sealed effectively, will allow  
128 magma to lose gas efficiently making an effusive eruption likely (Sparks, 1997). In  
129 comparison, a sealed system may lead to pressure build-up due to deep degassing and  
130 prime the volcano for an explosive eruption (Sparks, 1997; Komorowski et al., 1997;  
131 Burgisser et al. 2011; Preece et al., 2016; Roman et al., 2019; Komorowski et al., in prep).  
132 This indicates the role of bottom-up and top-down processes can cause transitions in  
133 eruptive behaviour both during a single eruption and across multiple eruptions.

134 In this study we focus on understanding the variability in explosive eruption style over  
135 several eruptions occurring within a period of ca. 7000 years. For this purpose, we study  
136 four eruptions from La Soufrière de Guadeloupe: the 5680 Cal. BCE Plinian eruption (VEI 4),  
137 the 1010 Cal. CE Plinian eruption (VEI 4), the 1530 Cal. CE sub-Plinian eruption (VEI 3) and  
138 1657 Cal. CE Vulcanian eruption (VEI 2). We use the chemical composition and volatile  
139 content of volcanic glass (melt inclusions and groundmass glass) to understand how the  
140 magmatic parameters such as, the pre-eruptive magma composition, conditions and  
141 physicochemical parameters in the conduit relate to variations in eruption styles.

## 142 **2 Geological and Magmatic Setting**

### 143 **2.1 Eruptive History**

144 Guadeloupe is found in the central Lesser Antilles arc and formed as a result of subduction  
145 of the Atlantic plate beneath the Caribbean plate. Basse-Terre, the western island of  
146 Guadeloupe, was formed over the last ca. 3 million years by the active inner volcanic arc  
147 which has produced several volcanic complexes (Komorowski et al., 2005; Samper et al.,  
148 2007). La Soufrière de Guadeloupe Volcano (hereby referred to as La Soufrière) is part of the  
149 most recently active Grande-Decouverte-Soufrière complex (Fig 1). Ca. 80,000 people live in  
150 the south of Basse Terre and would be impacted by a future eruption of La Soufrière  
151 eruption (Komorowski et al., 2005; Leone et al., 2019; Esposti Ongaro et al., 2020).

152 La Soufrière is a basaltic-andesite to andesitic arc volcano built over the older Monts  
153 Caraïbes complex and has been active since ca. 7140 Cal. BCE until present day (Legendre,  
154 2012; Komorowski et al., 2005; 2012; 2013). A large variation in magmatic eruption styles  
155 are observed at this system, from effusive dome building eruptions to explosive behaviour

156 ranging from Vulcanian to Plinian eruptions (Komorowski et al., 2005; Boudon et al., 2008;  
157 Legendre, 2012; Komorowski et al., 2013).

158 At La Soufrière two types of transitions between magmatic eruption styles are observed: (1)  
159 a transition in eruption style within the same eruption (e.g., 1530 Cal. CE which had  
160 explosive sub-Plinian and strombolian phases followed by an effusive dome building phase  
161 (Boudon et al., 2008; Komorowski et al., 2008), (2) a transition in eruption style over several  
162 eruptions with an observed repose period (e.g., the La Soufrière eruptive record shows  
163 individual eruptions separated a repose period ranging from Plinian to Vulcanian to effusive  
164 eruptions (Komorowski et al., 2005; Legendre, 2012).

165 Since 1635 Cal. CE, the majority of historic activity at La Soufrière has been phreatic  
166 (Feuillard et al., 1983; Komorowski et al., 2005), related to the well-developed hydrothermal  
167 system. Recent unrest at the system has suggested the movement and emplacement of  
168 magma at depth (Moretti et al., 2020) triggering pulses of recurrent heating and  
169 pressurisation of the hydrothermal system. However, there are few constraints on the  
170 controls of eruption style at this system and the transitions between eruption styles are  
171 poorly understood.

172 The eruptions presented in this paper cover the variety of explosive eruption styles which  
173 have occurred at La Soufrière through the Holocene, a full eruptive history is detailed in  
174 Komorowski et al. (2005); Legendre (2012) and Metcalfe et al. (2021). Four eruptions are  
175 discussed in detail: 5680 Cal. BCE, 1010 Cal. CE, 1530 Cal. CE and 1657 Cal. CE.

176 The 5680 Cal. BCE eruption is a Plinian eruption (VEI 4) and was the first explosive and most  
177 violent eruption preserved in the La Soufrière record. The 1010 Cal. CE eruption is the most  
178 recent Plinian eruption (VEI 4). The most well studied, archetypal eruption of La Soufrière is  
179 the multistage eruption of 1530 Cal. CE, that started with an edifice collapse phase, evolved  
180 into an explosive sub-Plinian (VEI 3) phase and ended with the growth of the current dome  
181 (Komorowski et al., 2005; Boudon et al., 2008; Komorowski et al., 2008; Esposti Ongaro et  
182 al., 2020). Finally, the last magmatic eruption was the 1657 Cal. CE a Vulcanian eruption (VEI  
183 2-3) (Komorowski et al., 2012; 2013; Legendre, 2012; Metcalfe et al., 2020). We also discuss  
184 basaltic samples from the Monts Caraïbes complex, which is found in the southernmost part  
185 of Basse-Terre and erupted between  $555 \pm 26$  and  $472 \pm 16$  ka (Blanc, 1983; Fig. 1). The least  
186 evolved magmas observed on Basse-Terre were produced during this phase, with the Monts  
187 Caraïbes magma often hypothesised as the parental magma from which other magmas on  
188 the island evolved (Bissainte, 1995).

189 Currently, La Soufrière has a developed hydrothermal system and following the 1657 Cal.  
190 CE eruption, only phreatic/hydrothermal eruptions have occurred, with the last event in  
191 1976-1977 (Feuillard et al., 1983; Komorowski et al., 2005). This eruption resulted in the  
192 evacuation of >70,000 people from Basse-Terre (Le Guern et al., 1980; Feuillard et al.,  
193 1983; Hincks et al., 2014) and resulted in increased monitoring of this system. The peak of  
194 recent unrest occurred in April 2018 when a M4.1 earthquake was recorded, this was the

195 strongest seismic event since 1976-1977. This is interpreted to relate to a failed phreatic  
196 event (Moretti et al. 2020).

## 197 **2.2 Magmatic System**

198 Models of the La Soufrière system involve the storage of a parental magma in a deep, zoned  
199 magma chamber below a system comprised of shallow-depth crystallised intrusions, as  
200 proposed by Touboul et al. (2007) and later by Pichavant et al. (2018). In both models the  
201 shallow andesitic system is remobilised by mafic magma moving up from the deeper  
202 system.

203 More recently, a mush system has been proposed for the structure of the La Soufrière  
204 magmatic reservoir (Moretti et al., 2020; Metcalfe et al., 2021). Mush systems are long-lived  
205 storage zones and are located at variable depths in the crust. These systems are composed  
206 of crystal and melt lenses in variable states of cooling and are built by repeated intrusions  
207 from depth (e.g., Mahood, 1990; Nakamura, 1995; Bachmann et al; 2002; Zellmer et al.,  
208 2003; Moretti et al., 2013, 2019; Brown et al., 2014; Cooper and Kent 2014; Annen et al.,  
209 2015; Bergantz et al., 2015, 2017; Edmonds et al., 2016; Cashman et al., 2017; Spera and  
210 Bohrson, 2018; Carrara et al., 2019). Crystals in the mush system have close contacts which  
211 form a continuous interlocking framework, through which melt is distributed in varying  
212 volumetric proportions and in variable states of eruptibility (e.g., varying temperature,  
213 density, viscosity, volatile content) (Cashman et al., 2017; Spera and Bohrson, 2018).

214 At La Soufrière, the mush lenses are hypothesised to vary through the reservoir with  
215 different lenses hosting different crystal populations (Metcalfe et al. 2021). This results  
216 from repeated intrusions interacting with the lenses differently due to the intrusion's size,  
217 geometry and location as well as physio-chemical properties (Metcalfe et al. 2021).

218 Melt and/or fluid migration, amalgamation and mixing render the system inherently  
219 unstable allowing remobilisation through disaggregation of the mush lenses (Costa et al.,  
220 2009; Mader et al., 2013; Rubin et al., 2017; Cooper and Kent, 2014; Sparks and Cashman,  
221 2017; Jackson et al 2018; Cooper 2019). At La Soufrière, it is hypothesised that magma  
222 recharge leads to remobilisation of magma mush (e.g., Bergantz et al., 2015, 2017;  
223 Schleicher et al., 2016; Schleicher and Bergantz 2017). Recharge may not result in  
224 remobilisation of the whole system, and only small areas may remobilise depending on  
225 intrusion geometry which relates to density differences between the intruding magma and  
226 host mush (Bergantz et al., 2015, 2017; Schleicher et al., 2016; Schleicher and Bergantz  
227 2017). This remobilised area may differ between eruptions, meaning not all the crystals  
228 interact with the same amount of intruding magma, therefore allowing different  
229 populations of crystals to be involved in different eruptions (Carrara et al., 2019; Cheng et  
230 al., 2020; Metcalfe et al. 2021).

231 The pre-eruptive melt composition is well constrained for the 1530 Cal. CE eruption  
232 (Poussineau, 2005; Boudon et al., 2008; Pichavant et al., 2018). Two magma compositions  
233 were involved to form the archetypal 1530 Cal. CE banded eruption products. This resulted

234 in a wide range of whole rock (WR) compositions observed from 55 - 62 wt% SiO<sub>2</sub> (Semet  
235 et al., 1981; Boudon et al., 2008; and Pichavant et al., 2018). However, the hybrid magma  
236 composition is not reflected in melt inclusions (MI), with inclusions from both mafic and  
237 silicic andesite bands falling in the range 70.5 – 78.7 wt% SiO<sub>2</sub> (Poussineau, 2005; Boudon  
238 et al., 2008 and Pichavant et al., 2018). Groundmass glass (GM) do reflect this hybrid  
239 composition, with the most silicic GM found in the homogenous andesitic pumice and  
240 white layers of the banded pumice, whereas more mafic glass is found in dark pumice  
241 bands. This indicates that the La Soufrière reservoir hosts an evolved melt composition and  
242 that recharge from depth, as in the case of the 1530 Cal. CE eruption, is more mafic with  
243 respect to the magma in the reservoir.

244 Quantification of MI concentrations and experimental data from 1530 Cal. CE MI studies by  
245 Pichavant et al. (2018) allows a pressure of 170 MPa for the source of this eruption to be  
246 constrained. This also allows a minimum depth for the top of the La Soufrière magma  
247 reservoir to be constrained between 5.6 and 7.1 Km. The absence of amphibole phenocrysts  
248 in comparison to other system in the Lesser Antilles also provides a maximum reservoir  
249 depth of 8.5 Km.

250 Glass studies by Pichavant et al. (2018) (MI) and Metcalfe et al. (2021) (GM) allow  
251 quantification of a range of temperatures for the system. The existing reservoir,  
252 hypothesised as an andesite body (Pichavant et al., 2018), is between 825 – 875°C. The  
253 recharging basalt was between 900–1025°C (Metcalfe et al., 2021) as it intruded into the  
254 system, and hybrid magmas, as observed in the 1530 Cal. CE eruption, are 925 - 950°C.

255 The Monts Caraïbes include the least evolved magmas observed on Basse-Terre and are  
256 often hypothesised as the parental magma from which other magmas on the island evolved,  
257 making it important to link the Monts Caraïbes to the La Soufrière magmas. The Monts  
258 Caraïbes is a large volcanic complex which began with submarine activity until the final  
259 phases which transitioned to sub-areal Plinian activity. Eruptive activity also produced an  
260 evolved dacitic composition as well as the basaltic component studied here (Bissainte, 1995;  
261 Komorowski et al., 2005).

### 262 **3 Methods**

263 Details on the sampling areas and methods used are available in Legendre, 2012, Metcalfe  
264 et al. 2021 and in the Supplementary Material provided. Juvenile clasts were collected from  
265 both proximal and distal locations from around the vent depending on where the freshest  
266 material was available from. Detailed sample descriptions, powder, mineral and melt  
267 inclusion preparation and analysis techniques can be found the Supplementary Material.

268 ICP-MS was conducted on whole rock samples to measure major element concentrations.  
269 Following sieving of the samples, juvenile pyroxene and plagioclase crystals were hand  
270 separated from the 500 µm sieved fraction and analysed at the mineral core and the rim.  
271 Melt inclusions are abundant in both pyroxene and plagioclase and are observed in all  
272 samples (Fig 2). For both pyroxene and plagioclase, the selected MIs had no evidence of



273 leakage or post-entrapment crystallisation (PEC) and were randomly distributed through the  
274 crystal. We also favoured crystals which hosted bubble-free inclusions in order to minimise  
275 the effect of volatile loss, especially CO<sub>2</sub> and S, to the vapour (e.g., Venugopal et al. 2020;  
276 Table S1). Further verifications were also carried out to ensure the MI can be interpreted as  
277 the original melt composition. We also observe Na-loss from both the MI and GM, despite  
278 analysing Na first.

279 EMPA was conducted on minerals and glass. Major and volatile compositions were obtained  
280 for 81 polished MIs, 35 hosted in pyroxene and 46 hosted in plagioclase across the different  
281 eruptions. SIMS analysis was conducted on 33 MIs to measure H<sub>2</sub>O, CO<sub>2</sub>, S, Cl and F  
282 concentrations. Calibration curves were established using data ranging from basaltic-  
283 andesite to rhyolite in composition (See supplementary material; Fig S1). Deuterium (D),  
284 expressed as D/H isotope ratio, was also measured in 28 representative MIs to assess the  
285 degree of H<sup>+</sup> diffusion which is used as a proxy for H<sub>2</sub>O loss (See supplementary material;  
286 Fig S2).

287 EMPA and SIMS analyses for S, Cl and F were compared and show a good agreement for S  
288 and Cl in particular. For example, for inclusion 1101viA-pyx-5 SIMS analysis 251 ppm S and  
289 1882 ppm Cl, in comparison EMPA analysis yielded 254 ppm S and 1720 ppm Cl. For F,  
290 however, there appears to have been some loss during EMPA analysis, for 1101viA-pyx-5  
291 SIMS analysis yielded 433 ppm F and EMPA analysis yielded 105 ppm F.

292 This difference in F may be due to the higher detection limit during EMPA analyses and the  
293 overlap of the the FK $\alpha$  peak by the FeL $\alpha$  peak in these relatively Fe-rich glasses (Todd 1996;  
294 Lowenstern, 1994; Witter and Kuehner 2004). For this reason, we use only the SIMS data for  
295 F.

## 296 **4 Results**

### 297 **4.1 Whole Rock**

298 Across Basse-Terre, bulk rock compositions from the various suites vary from basalt to  
299 dacite, with the least evolved compositions observed in the Monts Caraïbes suite in  
300 Southern Basse-Terre (Bissainte 1995; Boudon et al, 2008; Samper et al 2009; Fig 3). La  
301 Soufrière shows a narrower magma compositional range, from basalt to andesite (Boudon  
302 et al, 2008; Samper et al 2009).

303 This study provides further evidence that the Monts Caraïbes suite include the least evolved  
304 compositions observed on Basse-Terre showing a basaltic composition ranging from 47.6 to  
305 48.8 wt% SiO<sub>2</sub> for a range of eruption products including lava and scoria (Table S2; Fig 3).  
306 The younger La Soufrière magma suite from the 1657, 1010 and 1530 Cal. CE eruptions,  
307 show a basaltic-andesite to andesite composition (55.7 to 63.9 wt% SiO<sub>2</sub>; Fig 3), and so are  
308 more evolved than the older Monts Caraïbes magmas. Within the La Soufrière magmas,  
309 there is no clear compositional evolution with more evolved magma observed in 1530 Cal.  
310 CE (55.7 – 61.2 wt% SiO<sub>2</sub>) and 1010 Cal. CE (60.8 - 63.9 wt% SiO<sub>2</sub>) in comparison to the  
311 younger 1657 Cal. CE (57.9 – 62.1 wt%) samples.

312 The well-studied 1530 Cal. CE eruption has characteristic banding consistently observed in  
313 juvenile material with light bands are reported as ~60 wt% SiO<sub>2</sub> and less evolved dark bands  
314 with ~56 wt% SiO<sub>2</sub> (Fig 3; Boudon et al., 2008; Pichavant et al., 2018). Though the 1530 Cal.  
315 CE eruption shows a bimodal magma composition, the 1657 Cal. CE, 1010 Cal. CE and 5680  
316 Cal. BCE eruptions appear homogeneous with no compositionally banded material observed  
317 and relatively continuous whole rock compositions.

#### 318 **4.2 Groundmass Texture and Composition**

319 The groundmass textures are summarised in Table 1. The 1657 Cal. CE, 1010 Cal. CE and  
320 5680 Cal. CE GM are compositionally homogenous with differences observed in the  
321 microlite content (Fig 2). The 1530 Cal. CE GM is described by Pichavant et al. (2018),  
322 Boudon et al. (2008) and Martel et al. (2021) and varies between the light and dark layers  
323 of the banded pumices.

324 The GM compositions are more evolved than the whole rock compositions and fall in a  
325 relatively wide compositional range from andesite to rhyolite (Table S3; Fig 3). The older  
326 eruption (5680 Cal. BCE) extends to less evolved compositions: 61.1–75.8 wt% SiO<sub>2</sub>, 0.1– 3.6  
327 wt% MgO, 12.5 – 22.9 wt% Al<sub>2</sub>O<sub>3</sub>, 1.7 – 9.7 wt% FeO<sub>(total)</sub>, 3.1 – 8.9 wt% CaO and 0.6 – 1.9  
328 wt% K<sub>2</sub>O (Fig 4). In comparison, the younger eruptions (1010, 1530 and 1657 Cal. CE) show  
329 more evolved compositions: 60.6–76.3 wt% SiO<sub>2</sub>, 0.4– 4.7 wt% MgO, 11.5 – 18.4 wt% Al<sub>2</sub>O<sub>3</sub>,  
330 2.7 – 11.7 wt% FeO<sub>(total)</sub>, 2.1 – 7.4 wt% CaO, and 0.6 – 2.3 wt% K<sub>2</sub>O (Fig 4). Though there is a  
331 considerable overlap, the variations may relate to the abundance of microlites observed in  
332 the 5680 Cal. BCE eruption glass and not in the 1010 Cal. CE and 1657 Cal. CE.

333 We also observe a bimodal distribution in SiO<sub>2</sub> content for the 1657 CE eruption, with a high  
334 SiO<sub>2</sub> endmember (69.6 – 72.6 wt% SiO<sub>2</sub>) and a low SiO<sub>2</sub> endmember (60.6 – 67.6 wt% SiO<sub>2</sub>;  
335 Fig S3).

#### 336 **4.3 Mineral Texture and Composition**

337 The main mineral phases in all eruptions are plagioclase and pyroxene. The majority of  
338 pyroxene are orthopyroxene with small clinopyroxene populations observed in 5680 Cal.  
339 BCE and 1657 Cal. CE. Metcalfe et al. (2021), defines six different types of orthopyroxene in  
340 different eruptions of La Soufrière based on crystal textures and zoning patterns. MI are  
341 observed in all populations with no differences observed in the MI for different crystal  
342 types.

343 These populations, described in full in Metcalfe et al. (2021), include three simple crystals  
344 distinguished by their zoning patterns (Type-1: one sharp compositional zone; Type-2: one  
345 diffuse compositional zone; Type-3: multiple compositional zones), two complex crystals  
346 with resorption features (Type-4: resorption rims with remaining crystal core; Type-5:  
347 completely resorbed crystals) and unzoned crystals (Type-6: no compositional zones or  
348 variations). These six populations are also identified in this study and melt inclusions are  
349 observed in all six crystal types. Orthopyroxene compositions display a narrow

350 compositional range, particularly within the Type-1, 2, 3 and 6 crystals (En<sub>55-62</sub>). The type-4  
351 crystal cores also fall into this narrow compositional range, while the resorbed rims fall into  
352 a different compositional range which is the same as the type-5 resorbed crystals (En<sub>62-67</sub>).

353 The composition of pyroxene from the 1530 Cal. CE eruption is En<sub>56-59</sub>, which falls in the  
354 same compositional range observed for this study (Semet et al., 1981; Poussineau, 2005;  
355 Pichavant et al., 2018). Pichavant et al. (2018) also identify a small, sporadic population of  
356 Mg-rich orthopyroxene up to En<sub>71</sub>, which are closer to the resorbed compositions of type-4  
357 and type-5 crystals in this study. Bourgeoisat (2018) splits the 1530 Cal. CE orthopyroxene  
358 into three groups based on composition related to magmatic environment (ME): ME1 (En<sub>56-  
359 59</sub>), ME2 (En<sub>60-63</sub>), ME3 (En<sub>69-72</sub>). These compositions cover the range reported for this study  
360 with ME1 and ME2 compositions covering the range observed in the simple crystal Type-1,  
361 2, 3, 6 and 4 (cores) and ME3 are similar to the compositional range observed for Type-4  
362 (rims) and 5.

363 Plagioclase is present in all the eruptions discussed and the textures observed appear  
364 consistent through the eruptions. The plagioclase crystals are tabular and euhedral to  
365 subhedral in shape. Plagioclase often has oscillatory zoning and patchy zoning in the cores  
366 (Vance, 1965). Overall, the main populations in each eruption are approximately An<sub>60</sub> Ab<sub>40</sub>.  
367 Additional minor populations with different compositions are observed, notably an albite-  
368 rich population (An<sub>20-30</sub> Ab<sub>49-54</sub>) and a very anorthite-rich population (An<sub>80-94</sub> Ab<sub>6-19</sub>).

369 For the 1530 Cal. CE eruption, narrower plagioclase compositions are reported by  
370 Pichavant et al. (2018), with a main An<sub>60-65</sub> population. Anorthite-rich layers are also  
371 reported with An<sub>85-90</sub> (Semet et al., 1981; Pichavant et al., 2018). This suggests that the  
372 small populations of very anorthite-rich compositions observed in the eruptions from this  
373 study may correspond to the anorthite-rich layers reported by Semet et al. (1981) and  
374 Pichavant et al. (2018).

#### 375 **4.4 Melt Inclusions**

##### 376 **4.4.1 Post Entrapment Modifications**

377 Melt inclusions are droplets of silicate and form as melt surrounding a growing crystal  
378 becomes trapped in irregularities (Sorby, 1858; Cannatelli et al., 2016; Fig 4.2). Melt  
379 inclusions are composed of glass, dissolved volatiles, vapour bubbles and crystals in  
380 variable amounts and behave as a closed system once they have become trapped in the  
381 host crystal, so theoretically retain the melts original composition. A melt inclusion is After  
382 a melt inclusion (MI) has been trapped in a host crystal it may continue to evolve  
383 independent of the melt outside of the crystal (Roedder, 1984; Bodnar et al., 1989; Steele-  
384 Macinnis, 2011). The continued evolution of the MI means the composition no longer  
385 represents the composition of the melt which was trapped. For example, crystallisation of  
386 daughter minerals or along the MI wall and diffusive re-equilibration between the MI and  
387 host crystal can all result in compositional changes (Gaetani and Watson, 2000; Schiano,  
388 2003; Wallace, 2005; Steele-Macinnis et al., 2011). Volatiles, including CO<sub>2</sub> which have a

389 low solubility in the melt, can be lost in significant percentages to vapour bubbles  
390 (Anderson and Brown 1993; Venugopal et al., 2020).

391 H<sub>2</sub>O loss from the MI can occur if the host crystal fractures or due to H<sup>+</sup> diffusion through  
392 the host crystal (Anderson, 1974a; Hauri, 2002; Danyushevsky et al., 2002; Wallace, 2005).  
393 The possibility of H<sub>2</sub>O loss makes it is necessary to assess if observed H<sub>2</sub>O trends are the  
394 result of trapping of variably degassed melts or due to H<sup>+</sup> loss. To assess H<sup>+</sup> diffusion the  
395 D/H isotopic ratio can be measured, this is expressed as  $\delta D$  (the permil deviation from  
396 Standard Mean Ocean Water,  $\delta D_{SMOW}$ ). For slab fluids associated to the average upper  
397 MORB mantle,  $\delta D$  typically falls in the range of -20 to -80 ‰ (Shaw et al., 2008). Loss of H<sup>+</sup>  
398 and addition of seawater and/or secondary hydrothermal fluids is also shown by positive  $\delta D$   
399 values and negative correlations with H<sub>2</sub>O (wt%) (Kyser and O'Neil, 1984; Shaw et al., 2008;  
400 Portnyagin et al., 2008). Post-entrapment modifications (PEM) can also be assessed by  
401 looking at the equilibrium between the MI and host crystals (Putirka, 2008). If the host  
402 crystal and MI are in equilibrium it can be assumed that the MI has experienced minimal  
403 post-entrapment processes such as diffusive loss and crystallisation and is representative of  
404 the original magma composition.

405 This included examining H<sup>+</sup> loss, for the eruptions discussed here, minimal H<sup>+</sup> loss is  
406 observed for most inclusions Generally, most inclusions fall into the D/H isotopic ratio  
407 range, expressed as  $\delta D$  -25 to -70‰ (Fig S2). One very positive value suggesting H<sup>+</sup> loss is  
408 observed for 1657 Cal. CE and one very negative value is also observed for 5680 Cal. BCE,  
409 which is due to the inclusions in this sample being small. However, as most inclusions are in  
410 the range for typical slab fluids (-20 to -80 ‰; Shaw et al., 2008), we assume the H<sub>2</sub>O  
411 contents are representative of the magma.

412 Post-entrapment modification of the melt inclusions, especially the host mineral  
413 crystallisation at the walls of the MI, was assessed in two ways: (1) the MI compositions  
414 were compared to groundmass glasses and whole rock compositions in order to identify  
415 anomalous compositions with respect to the La Soufrière liquid line of descent. Though  
416 some Na<sub>2</sub>O loss during analysis is observed, MI fit into the liquid line of descent (Fig 3 and  
417 4). (2) Using the method of Putirka, 2008, the MI-host crystal equilibrium ( $K_D$ ) was assessed  
418 and only MI in equilibrium are included in the final data set. For pyroxene hosted MIs and  
419 host crystal pairs the  $K_D$  is calculated by comparing the Mg/Fe ratios in the MI and host  
420 crystal (Putirka, 2008). This was also used for groundmass glass and pyroxene crystal pairs  
421 (Metcalf et al. 2021).  
422 MI with a  $K_D$  in the range 0.23 – 0.35 (Putirka, 2008) are considered to be in equilibrium and  
423 can be used in the dataset. The equilibrium values calculated for the pyroxene MI fall into  
424 the 0.23 – 0.35 range, with any inclusions falling outside this range excluded from the  
425 dataset (Table 2).

426 For plagioclase hosted MIs and host crystal pairs the  $K_D$  is calculated by comparing the albite  
427 (Ab) and anorthite (An) content of the host crystal with the Al, Ca, Na and Si content of the  
428 MI (Putirka, 2008). For plagioclase the equilibrium values are temperature dependent: MI

429 trapped at <1050°C are in equilibrium in the range 0.05 – 0.15, and MI trapped at >1050°C  
430 are in equilibrium in the range 0.17-0.39 (Putirka, 2008). The majority of plagioclase MI  
431 record temperatures of <1050°C and  $K_D$  in the range 0.05 – 0.15. MI outside of this range  
432 were excluded from the study. The few MI recording temperatures >1050°C have  $K_D$  in the  
433 range 0.17-0.39 (Table 2).

434 By only including MI in the equilibrium ranges ensures all MI discussed in this paper have  
435 experienced minimal post-entrapment modifications. This includes the 1530 CE MI which  
436 are described by Pichavant et al. (2018) as having experienced no significant post-  
437 entrapment modification (See Supplementary Fig S4).

#### 438 **4.4.2 Major and Volatile Elements**

439 The pyroxene MI are found across a range of crystal types described in Metcalfe et al., 2021  
440 (Table S1). MIs of the 1657 and 1010 Cal. CE eruptions are all found in the cores and rims of  
441 simple orthopyroxene crystals (Type 1-3 and 6), while the MIs of the 5680 Cal. BCE eruption  
442 are found in cores and rims of both simple and complex orthopyroxene crystals. However,  
443 no difference in MI composition is observed between the different crystal types.

444 MIs of the 5680 Cal. BCE eruption are dacitic in composition (64.6 - 76.7wt% SiO<sub>2</sub>; see  
445 supplementary Fig S5). The majority of the MIs of the 1010 Cal. CE eruption (64.3 - 77.5 wt%  
446 SiO<sub>2</sub>; see supplementary Fig S6) and 1657 Cal. CE eruption (73.1 – 78.2 wt% SiO<sub>2</sub>; see  
447 supplementary Fig S6) are also dacitic in composition with few MIs of rhyolitic in  
448 composition (Fig 4; Table 2). The majority MIs of the 1530 Cal. CE eruption are reported to  
449 be rhyolitic in composition with a small population which are dacitic in composition (70.5-  
450 78.7 wt% SiO<sub>2</sub>; see supplementary Fig S6) (Poussineau, 2005; Boudon et al., 2008; Pichavant  
451 et al., 2018). There is a relatively large compositional range, however, this remains relatively  
452 constant across the eruptions studied: 64.3– 78.7 wt% SiO<sub>2</sub>, 0.02– 2.8 wt% MgO, 11.9 – 21.3  
453 wt% Al<sub>2</sub>O<sub>3</sub>, 0.6 – 8.2 wt% FeO<sub>(total)</sub>, 1.1 – 7.0 wt% CaO, 0.05 – 4.7 wt% Na<sub>2</sub>O and 0.7 – 2.7  
454 wt% K<sub>2</sub>O (Fig 4; Table 2).

455 For all the eruptions, with increasing SiO<sub>2</sub> contents CaO, FeO, MgO, MnO, Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and  
456 Na<sub>2</sub>O decrease and K<sub>2</sub>O increases. MIs, along with the groundmass glass, exhibit linear  
457 trends (Fig. 4; Table 2). The compositions of the MIs also show considerable overlap with  
458 the groundmass compositions. For the 1010 Cal. CE and 5680 Cal. BCE eruptions most MI  
459 are more evolved than the groundmass glass compositions, in comparison the 1657 Cal. CE  
460 eruption where MI are generally the same composition or less evolved than groundmass  
461 glass. The MIs of the 1530 Cal. CE eruption and groundmass glass compositions are very  
462 similar, with inclusions varying between being more and less evolved than glass  
463 compositions (Boudon et al., 2008, Poussineau, 2005; Pichavant et al., 2018).

464 We also observe variations that distinguish between the VEI 4 eruptions and the VEI 2-3  
465 eruptions when looking at the average MI SiO<sub>2</sub>. The average MI SiO<sub>2</sub> (wt%) is lower for both  
466 the VEI 4 eruptions (1010 CE – 71.6 wt% SiO<sub>2</sub>, 5680 BCE – 72.7 wt% SiO<sub>2</sub>) in comparison to

467 the VEI 2-3 eruptions (1657 CE – 76.1 wt% SiO<sub>2</sub> and 1010 CE – 74.3 wt% SiO<sub>2</sub>). Giving a  
468 compositional gap of 1.6 - 4.5 wt% SiO<sub>2</sub> between the two VEI 4 and VEI 2-3 eruptions.

469 The range of water across the eruptions is 1.3 - 4.4 wt%, with little variation in the water  
470 content between eruptions (5680 Cal. BCE: 2.4 - 4.2 wt%; 1010 Cal. CE: 1.3 - 4.4 wt% and  
471 1657 Cal. CE: 2.3 - 4.1 wt% (Fig 5A; Table 2)). The reported range for the 1530 Cal. CE  
472 eruption is 2.5 - 5.6 wt% H<sub>2</sub>O, however, most inclusions are reported to have 5 - 5.5 wt%  
473 H<sub>2</sub>O which is higher than recorded for the eruptions in this study (Pichavant et al., 2018) (Fig  
474 5A; Table 2).

475 In comparison to H<sub>2</sub>O contents, CO<sub>2</sub> contents vary greatly across the eruptions. The range  
476 measured in bubble-free MI across the eruptions is 11 – 866 ppm. The highest CO<sub>2</sub> contents  
477 are recorded for the 5680 Cal. CE eruption with a range from 116-866 ppm. The lowest CO<sub>2</sub>  
478 contents are recorded for the 1010 Cal. CE eruption with CO<sub>2</sub> contents often below the  
479 detection limit and a range from 11 – 405 ppm. The CO<sub>2</sub> contents for the 1657 Cal. CE  
480 eruption ranges from 35 - 674 ppm. Across the eruptions CO<sub>2</sub> contents decrease with  
481 increasing K<sub>2</sub>O (Fig 5B; Table 2). No CO<sub>2</sub> contents have been published for 1530 Cal. CE.

482 Sulphur (S) is relatively consistent across the eruptions, ranging from 20-777 ppm. There is  
483 little difference S content between the eruptions (e.g., 5680 Cal. BCE: 20-777 ppm; 1010 Cal.  
484 CE: 57-654 ppm and 1657 Cal. CE: 30-564 ppm. The S contents reported for 1530 Cal. CE  
485 (184-495 ppm) by Poussineau (2005) and Pichavant et al. (2018) also fall in this range (Fig  
486 5C; Table 2).

487 Chlorine (Cl) has a range of 892 - 3610 ppm across the eruptions. Similar to S, there is little  
488 variation in the Cl content between eruptions (e.g., 5680 Cal. BCE: 1041 – 2551 ppm; 1010  
489 Cal. CE: 1458-3198 ppm and 1657 Cal. CE: 892 – 3610 ppm (Fig 5D; Table 2). The Cl contents  
490 reported for 1530 Cal. CE (2462- 3006 ppm) by Poussineau, 2005 and Pichavant et al., 2018  
491 also fall in this range.

492 Fluorine (F) content ranges from 350 to 610 ppm across the eruptions (only SIMS data). The  
493 F data shows little variation between eruptions (5680 Cal. BCE: 370 - 610 ppm; 1010 Cal. CE:  
494 350 - 483 ppm and 1657 Cal. CE: 400-510 ppm (Fig 5E; Table 2)). No F contents have been  
495 published for 1530 Cal. CE.

496 The total volatile content across the eruptions is relatively consistent with the majority of  
497 the total volatiles made up by H<sub>2</sub>O (Fig 5F; Table 2): 5680 Cal. BCE - 4.7 wt%; 1010 Cal. CE -  
498 5.1 wt%; 1530 Cal. CE - 5.3 wt%; 1657 Cal. CE - 4.7 wt%.

## 499 **5 Discussion**

### 500 **5.1 The Mush Model of La Soufrière**

#### 501 **5.1.1 Magmatic Conditions**

502 The results of our MI analysis allow us to incorporate a more accurate understanding of  
503 temperatures, pressures, depths and melt compositions into the mush model proposed by

504 Metcalfe et al. (2021) for La Soufrière. Scandone et al. (2007) schematise four regions to  
505 describe the generation and ascent of magma in a general context: (i) the deepest part of  
506 the system: the supply system, (ii) the intermediate storage: the mush zone, (iii) the  
507 conduit, (iv) the vent. The four regions are not necessarily simultaneously connected  
508 depending on the supply rate from the supply system and processes occurring in the mush  
509 storage zone. Here we use a similar schematic and our study focuses on the mush storage  
510 zone. This region is formed from repeated intrusions from the supply system and is  
511 composed of an interlocking framework of crystals which melt is distributed through the  
512 mush in varying volumetric proportions. Melt and/or fluid migration, amalgamation and  
513 mixing make the system inherently unstable allowing remobilisation for example, by  
514 pressure increase from crystallisation or influxes of new magma (Scandone et al., 2007;  
515 Costa et al., 2009; Mader et al., 2013; Moretti et al., 2013; Brown et al., 2014; Rubin et al.,  
516 2017; Cooper and Kent, 2014; Sparks and Cashman, 2017; Jackson et al 2018).

517 The MIs show that the composition of the La Soufrière magma has remained relatively  
518 stable across the eruptions studied here, and no compositional evolution across the history  
519 of the system is observed (Fig 4). This suggests that the composition of any new magma  
520 inputs has also remained relatively similar. However, as the MI from the La Soufrière  
521 eruptions show an evolved composition, these MI do not preserve the primitive melt  
522 composition feeding this system. Based on this, we can only make inferences about the  
523 mush storage region and not on deeper processes occurring in the supply zone involving  
524 more primitive melts.

525 The MI and GM relationship provides further evidence of a mush storage zone at La  
526 Soufrière (Fig 4). The fact that some MI have a more evolved composition than the GM  
527 indicates the MI have cooled prior to interaction with the GM and are exotic to the  
528 transporting melt (e.g., Kilgour et al., 2013). This is possible if the transporting melt entrains  
529 MI-bearing crystals stored in the mush system. The 5680 Cal. BCE, 1010 Cal. CE, 1530 Cal. CE  
530 and 1657 Cal. CE eruptions all have MI which are more evolved than the GM glass, indicating  
531 MIs have been entrained from the mush lenses in the erupting melt. This could be  
532 particularly important for the 5680 Cal. BCE eruption where orthopyroxene crystals are  
533 observed to have resorption rims (Metcalfe et al., 2021).

534 The MIs can also be used to calculate the properties of the mush storage zone. The  
535 thermobarometers of Putirka (2008) were used to calculate the temperatures for both MI  
536 (Putirka, 2008 eq. 24a for plagioclase, eq. 28a for orthopyroxene and eq. 33 for  
537 clinopyroxene) and groundmass glass (with orthopyroxene rims, Putirka, 2008: eq. 28a;  
538 Metcalfe et al. 2021), this thermobarometer has an error of  $\pm 30^{\circ}\text{C}$ . For each eruption, MI  
539 record a range of temperatures: 965 – 1110 $^{\circ}\text{C}$  for 5680 Cal. BCE, 960 – 1050 $^{\circ}\text{C}$  for 1010 Cal.  
540 CE, and 945 – 1050 $^{\circ}\text{C}$  for 1657 Cal. CE (Fig 6A). GM record a range of temperatures: 870 –  
541 1085 $^{\circ}\text{C}$  for 5680 Cal. BCE, 1010 – 1025 $^{\circ}\text{C}$  for 1010 Cal. CE, and 950 – 1020 $^{\circ}\text{C}$  for 1657 Cal. CE  
542 (Fig 6A). The temperatures reported by Pichavant et al., (2018) for 1530 Cal. CE (825-  
543 1025 $^{\circ}\text{C}$ ) are in a similar range as those calculated for this study. (Fig 6A). For the 1657 CE,

544 1010 CE and 5680 BCE eruptions there is significant overlap between the mean MI (1000  
545  $\pm$ SD 40 °C, 1000  $\pm$ SD 35 °C and 1020  $\pm$ SD 36 °C, respectively) and GM temperatures (975  $\pm$ SD  
546 24 °C, 980  $\pm$ SD 8 °C and 990  $\pm$ SD 72 °C, respectively).

547 Using the method of Papale et al. (2006) minimum pressures were calculated using the  
548 highest H<sub>2</sub>O and CO<sub>2</sub> contents recorded by MIs for each eruption. The ferric and ferrous iron  
549 con- tents of the melt were estimated using Kress and Carmichael (1991) which is integrated  
550 into MagmaSat (Ghiorso and Gualda, 2015). This shows the pressure of MI entrapment  
551 ranges from 190 MPa to 220 MPa (Fig 6B). These pressures are similar to the pressure of  
552 170 MPa reported by Pichavant et al. (2018) for the 1530 Cal. CE eruption (Fig 6B).  
553 Converting the pressures to depths using a density of 2450 kg m<sup>-3</sup> (Barnoud et al., 2016)  
554 indicates that these eruptions at La Soufrière were fed by melts originating from minimum  
555 depths of ~7 to 9 km within the mush storage zone. However, different eruptions were fed  
556 by melt originating from different areas of the mush storage zone and the 5680 Cal. BCE  
557 eruption was fed from the deepest source at ~8.7 Km. This shows the eruptions of La  
558 Soufrière studied here have been fed from a narrow range of depths within the mush  
559 system.

560 Oxygen fugacity ( $fO_2$ ) affects magma phase equilibria, density and rheology because of  
561 redox reactions affecting fluid speciation and iron speciation in melts (Kress and Carmichael,  
562 1991; Campbell et al., 2009; Moretti and Papale, 2004; Burgisser and Scaillet, 2007; Moretti  
563 and Stefansson, 2020). In this study, we adopt the Kress and Carmichael (1991) equation,  
564 which is included in the Ghiorso and Sack (1995) model for natural magmas' phase  
565 equilibria. This allows  $fO_2$  to be defined from a measured melt composition and calculated  
566 temperature and pressure. For La Soufrière, this model shows the  $\log fO_2$  ranges from -9.1 - -  
567 9.4. For each eruption the average  $\Delta NNO$  values are: +0.86 (5680 Cal. BCE), +0.79 (1010 Cal.  
568 CE) and +0.85 (1657 Cal. CE), calculated using the average melt inclusion temperatures  
569 (1020°C, 1000°C and 1000°C, respectively). These values are in the same range reported for  
570 1530 Cal CE ( $\Delta NNO$ +0.8) by Pichavant et al. (2018) and indicates that the  $fO_2$  of melts at La  
571 Soufrière has remained relatively constant through time.

### 572 **5.1.2 Magmatic Processes at Depth**

573 Understanding the processes occurring in the storage zone is important to understand the  
574 dynamics, chronology and style of eruptions. Pichavant et al. (2018) use melt inclusions and  
575 experimental data to investigate the processes driving the 1530 Cal. CE eruption. For this  
576 eruption, the andesite in the storage zone is remobilised by the arrival of a less evolved  
577 basaltic magma. Mixing between the andesite and basalt forms a hybrid magma where  
578 banded material is extracted from. The mixing is incomplete with an unmixed andesite,  
579 hybrid magma and unmixed basalt present. In this scenario, recharge by a less evolved  
580 magma from depth is needed to explain the observed eruption products and compositions.

581 The eruptions detailed in the present work, in contrast to 1530 Cal. CE, do not show any  
582 textural evidence of mixing with more primitive compositions which may have triggered the



583 eruption. However, for the 1657 CE eruption there is some evidence for a bimodal  
584 distribution within the groundmass glass compositions which could indicate incomplete  
585 mixing of high and low SiO<sub>2</sub> melts (Fig S3). This suggests both the 1657 Cal. CE and 1530 Cal.  
586 CE eruptions may have been driven by mafic recharge intruding into the storage zone.

587 For the 1010 Cal. CE and 5680 Cal. BCE eruptions, the lack of compositional and textural  
588 evidence for mixing could indicate that recharges from the supply system have not been  
589 involved in these eruptions. Alternatively, efficient mixing could have occurred, or magmas  
590 preserving mixing textures have not been erupted. In this case, mafic recharge would  
591 prompt the reactivation of shallow magmas by supplying it with fluids and heat, as for other  
592 volcanoes of Lesser Antilles (e.g., Edmonds et al., 2010; Solaro et al., 2020).

593 Furthermore, the occurrence of potential mafic recharge may explain the lack of  
594 compositional evolution across the samples, with the compositions related to the volumes  
595 of mafic/evolved endmembers and the efficiency of magma mixing. Alternatively, the fact  
596 that not all parts of the mush system may be tapped during eruption would again contribute  
597 to the lack of any broad compositional evolution (Metcalf et al., 2021).

598 The compositional gap between the VEI 4 eruptions and the VEI 2-3 eruptions could suggest  
599 the Plinian events erupted well-mixed, moderately evolved magmas. In comparison, the  
600 lower magnitude eruptions erupted poorly mixed magmas with the MI representing the  
601 mush-zone magma and evidence for mixing observed in groundmass glass compositions and  
602 textures. Poorly mixed magmas are widely recorded for VEI 2-3 eruptions at arc volcanoes,  
603 including: Turrialba (DeVitre et al., 2019); Santiaguito (Wallace et al., 2020); Tungurahua  
604 (Myers et al., 2014); Mt Unzen (Nakamura, 1995); Kasatochi (Neill et al., 2015); Soufriere  
605 Hills, Montserrat (Barclay et al., 2010; Edmonds et al., 2010) and Kelud (Jeffery et al., 2013).

606 We also consider the possibility that a more evolved melt migrated through the mush zone  
607 following the 1010 CE eruption, and that subsequent mafic injections interacted and began  
608 to mix and mingle with the more evolved melt to produce to 1530 CE (very poorly mixed)  
609 and 1657 CE (moderately poorly mixed) eruption products.

## 610 **5.2 Controls on Eruption Style**

611 The overall stability of the temperature and depth (7 - 9 Km) indicates the storage zone has  
612 remained relatively stable over time. Though the eruptions studied here have been fed from  
613 a relatively narrow range of depths, we cannot rule out that the mush system may extend  
614 over a larger range of depths. Although the composition and volume of any potential mafic  
615 recharge may also result in variation in eruption style, this suggests that factors and  
616 processes occurring above the magma storage zone and within the conduit system may also  
617 play a considerable role in controlling eruption style. These processes include degassing of  
618 the ascending magmas, rheological changes and the interactions with other top-down  
619 processes of non-magmatic but external origin. Particularly in a mushy system, these  
620 processes modulate to various extents the magma ascent rate, which initially is ideally

621 imposed by the mass flux at the exit of the magma storage zone. Therefore, though  
622 eruption style may be determined by factors and processes occurring above the magma  
623 storage zone, and not influenced by dynamics in the magma storage zone, the magma  
624 storage zone must still be considered to play a role in driving the build-up to an eruption.

### 625 **5.2.1 Volatiles**

626 Volatiles are considered to be an important control on eruption style (Blundy and Cashman,  
627 2005; Dingwell, 1996; Melnik and Sparks, 2002). Volatiles, in particular H<sub>2</sub>O, have an  
628 important control on magma ascent. Degassing during ascent leads to degassing-induced  
629 crystallisation (Cashman and Blundy, 2000; Cashman, 2004) which promotes further volatile  
630 exsolution and results in changes in magma rheology (Dingwell, 1996; Edmonds and  
631 Wallace, 2017). Alternatively, exsolution-driven volatile expansion drives fast ascent which  
632 results in high volatile overpressure in bubbles and high strain rates from rapid acceleration  
633 leading to explosive fragmentation (Roggensack et al., 1997; Papale, 1999; Huppert and  
634 Woods, 2002; Scaillet et al., 2008; Owen et al., 2013; Cassidy et al., 2016; Bernard and  
635 Bouvet de Maisonneuve, 2020).

636 In the case of La Soufrière, it could be expected that the large magnitude eruptions of  
637 Plinian style were driven by higher H<sub>2</sub>O contents. However, no discernible differences are  
638 observed, with the large 5680 Cal. BCE and 1010 Cal. CE Plinian eruptions and the 1657 Cal.  
639 CE Vulcanian eruption having <0.3 wt% difference in H<sub>2</sub>O content.

640 Open system degassing and fluxing of CO<sub>2</sub> are important process at some systems to drive  
641 explosive eruptions (Sable et al., 2006; Aiuppa et al., 2011; Blundy et al., 2010; Moretti et  
642 al., 2019; Assbichler, 2020; Allison et al., 2021). However, the plot of H<sub>2</sub>O versus CO<sub>2</sub> shows  
643 no clear degassing trends, instead the wide spread of H<sub>2</sub>O and CO<sub>2</sub> contents indicates  
644 complex degassing histories and the trapping of variably degassed melt (Schmitt 2001; Atlas  
645 et al. 2006; Johnson et al. 2010; Blundy et al. 2010; Reubi et al., 2013; Fig S8).

646 The fluxing of CO<sub>2</sub> occurs because deep melts with high CO<sub>2</sub> contents degas and supply CO<sub>2</sub>  
647 to shallow ascending magmas but it seems to control the eruption style wherever the total  
648 (exsolved + dissolved) CO<sub>2</sub> amount is high (>0.6. wt%, Allison et al., 2021; > 1.5 wt% in most  
649 cases, Blundy et al., 2010; Moretti et al., 2018, 2019). Current data do not allow total CO<sub>2</sub> in  
650 the system to be assessed reliably (e.g., Barsanti et al., 2009; Moretti et al., 2018), but based  
651 on degassing and fluxing simulations carried out on Montserrat volcanoes total CO<sub>2</sub> is not  
652 expected to be larger than 0.2 wt%, inferred from the amount dissolved in the unerupted  
653 underplating basalt (Edmonds et al., 2010). Considering that CO<sub>2</sub> contents measured in this  
654 study for La Soufrière are even lower (<0.1 wt%), which is also the case for Montserrat  
655 basalt values, we can conclude the total CO<sub>2</sub> content is not a major control, with H<sub>2</sub>O  
656 making up the majority of the total volatile content.

657 Assuming that the bulk dissolved content of the most primitive MIs approximate the total  
658 volatile content of magmas in their subsequent evolution toward pre-eruptive conditions,  
659 there is <1 wt% difference in total volatile content across the eruptions studied, suggesting

660 the volatile content cannot be the main controlling parameter for eruption style (Fig 5F).  
661 Comparing the total volatile content against VEI highlights that, for these eruptions, volatile  
662 content does not control explosivity (Fig 5F). However, we cannot discount the effect of an  
663 exsolved fluid phase on the eruption style. At La Soufrière, gas measurements currently  
664 acquired over the active fumarolic field allow to quantify the degassing volatile flux.  
665 However, the lack of such data for past eruptions to compare with our MI dataset  
666 significantly complicates the understanding of the exsolved fluid phase. Further to this,  
667 current measurements of the gas phase composition at the surface do not reproduce the  
668 pre-eruptive magmatic volatile phase at depth because the well-developed hydrothermal  
669 system scrubs the magmatic signal (Symonds et al., 2001; Moretti et al., 2020; Moune et al.,  
670 2022).

### 671 **5.2.2 Viscosity Changes**

672 To investigate how the magma has changed during ascent and eruption we investigate the  
673 syn-eruptive change of flow behavior. Crystallization in the conduit can abruptly modify  
674 magma properties and effect eruption style (Gardner et al., 1998; Nakada et al., 1999;  
675 D’Oriano et al., 2005; Di Genova et al., 2017; Hlinka, 2020; Romano et al., 2020; Vona et al.,  
676 2020; Hlinka et al., 2021). As the MI are trapped in phenocrysts formed in the storage zone,  
677 we instead use GM viscosity where the microlite crystallization relates to changes occurring  
678 in the conduit during magma ascent prior eruption (Hlinka, 2020; Hlinka et al., 2021). We  
679 assume crystallization of the GM occurred in the conduit and was quenched on eruption  
680 and no further crystallization occurred after eruption.

681 The syn-eruptive crystal fraction in the GM varies for each eruption (Table 3), with the  
682 largest variation observed between the 5680 Cal. BCE microlite-rich Plinian eruption and the  
683 1010 Cal. CE microlite-poor Plinian eruption. The ConFlow 1.0.5 model (Mastin, 2002) is  
684 used to calculate the groundmass viscosity as this model takes into consideration  
685 crystallinity. Using the ConFlow 1.0.5 model (Mastin, 2002) crystal-free viscosity model,  
686 mean glass composition, magma temperature and pressure we calculate GM viscosities  
687 ranging from  $10^{5.4}$  Pa·s to  $10^{8.2}$  Pa·s (Table 3; Fig 6C). Notably, the two Plinian eruptions  
688 (5680 Cal. BCE and 1010 Cal. CE) are characterised by lower viscosities than the two  
689 smaller eruptions (1530 Cal. CE and 1657 Cal. CE) (Fig 6C). This is one of the only  
690 properties which shows a clear difference between the more explosive (VEI 4) and less  
691 explosive (VEI 2-3) eruptions.

692 Using the microlite vol%, mean glass composition and the ConFlow 1.0.5 model (Mastin,  
693 2002) we calculate the melt viscosity after syn-eruptive crystallisation (GM+X) (Figure 6C).  
694 The resulting increase in the effective viscosity between GM and GM+X indicates an  
695 increase in potential for explosivity for the 1657 Cal. CE, 1530 Cal. CE and particularly, the  
696 5680 Cal. BCE eruptions (e.g., Houghton and Gonnermann, 2008; Moitra et al., 2018; Arzilli  
697 et al., 2019; Hlinka, 2020; Hlinka et al., 2021). The range of viscosities calculated for La  
698 Soufrière have a similar range to viscosities calculated in the literature (See Supplementary  
699 Fig S9), which for groundmass glass without microlites range from  $10^{1.9}$  -  $10^{8.2}$  Pa·s and for

700 groundmass glass with microlites range from  $10^2 - 10^{9.7}$  Pa·s (Taddeucci, et al., 2004; Lautze  
701 and Houghton, 2007; Pappalardo et al., 2018; Mujin and Nakamura, 2020; Hlinka et al.,  
702 2021).

703 The 5680 Cal. BCE eruption experienced a larger viscosity increase than the 1657 and 1530  
704 Cal. CE eruptions, suggesting the magma feeding the 5680 Cal. BCE eruption had a higher  
705 explosivity following ascent. This large viscosity change may have resulted in the 5680 Cal.  
706 BCE eruption being the largest Plinian eruption in the La Soufrière record. However,  
707 viscosity changes cannot be the only cause of Plinian eruptions at this system, as a large  
708 viscosity change is not observed for the magma feeding the 1010 Cal. CE Plinian eruption.  
709 For the 1010 Cal. CE eruption a combination of other factors may result in the culmination  
710 of a Plinian eruption.

### 711 **5.2.3 Ascent Rate**

712 The ascent rate is the rate which magma ascends through the conduit from the magma  
713 mush storage zone to the surface and is a function of pressure magma density, viscosity,  
714 and resistance to flow while in the conduit (Papale and Dobran 1994; Mastin and Ghiorso  
715 2001; Pinkerton et al. 2002; Sparks et al. 2006; Rutherford, 2008). Ascent rate has been  
716 shown to be an important factor in controlling the onset of fragmentation, hence the  
717 transition between eruption styles and so may also be a key parameter to explain the  
718 differences in eruption styles observed at La Soufrière (e.g., Cassidy et al., 2018).

719 Here we simplify magma ascent to fast and slow relative to the rates calculated. The ascents  
720 calculated refer to an average velocity from the storage system to the surface, though in  
721 reality magma ascent rate will vary through the conduit due to changes in vesicularity and  
722 volatile content, overpressure at depth, magma rheology and conduit geometry.

723 ConFlow 1.0.5 from Mastin (2002) is used to calculate ascent rate based on crystal volume  
724 % and mean GM composition. The model also requires the input of temperature, pressure  
725 and depth (taken from Putirka, 2008 and Papale et al., 2006 calculations). Conduit diameter  
726 is also required when using this model and is the largest unknown as no magmatic eruptions  
727 have been observed at La Soufrière so estimates must be used.

728 The conduit is defined as the connection between the magma storage area and the surface  
729 (Wilson et al., 1980; Bower and Woods, 1996). In explosive eruptions at more evolved  
730 systems the conduit is hypothesised to begin as a small fracture which widens allowing  
731 higher velocities, resulting in erosion of the conduit and widening into the vent (Wilson et  
732 al., 1980). To simplify the model, the conduit is assumed to be a vertical cylinder with a  
733 constant radius from the storage area to the surface which does not include any widening in  
734 the upper parts of the conduit or the vent radius (Fedotov, 1981; Colucci et al., 2014).

735 One way to estimate conduit diameter is to look at similar systems. At the neighbouring  
736 volcano Soufrière Hills Volcano, Montserrat, a conduit radius of 15 m has been estimated  
737 for the sub-Plinian eruption on 17<sup>th</sup> September 1996 (Robertson et al., 1998). This is based  
738 on Bower and Woods (1996) which models a sustained discharge from a narrow conduit

739 into a wide crater, from the initial crater dimensions, spine widths from earlier eruption  
740 phases and fluid dynamic models (Robertson et al., 1998). A 15 m radius is also estimated  
741 for smaller Vulcanian eruptions occurring at Soufrière Hills Volcano during August 1997  
742 where a 30 m  $\pm$ 5 m diameter conduit, widening into a flared crater was estimated using  
743 spine dimensions, magma ascent rates and volume extrusion rates (Voight et al., 1999;  
744 Clarke et al., 2002). A radius of 15 m has also been used for modelling the dynamics of lava  
745 dome extrusion during the 1995-1999 period (Devine et al., 1998; Melnik and Sparks, 1999).  
746 Based on this we used conduit diameters of 10, 20, 30, 40 m to cover the observed conduits  
747 at Montserrat. ConFlow 1.0.5 assumes the conduit is vertical and circular in cross section.

748 For 10 to 40 m diameters, this model gives ascent rates of 0.7 – 3 m/s for the 1657 Cal. CE  
749 Vulcanian eruption and 2 – 4 m/s for the 1530 Cal. CE sub-Plinian eruption (using the  
750 average microlite vol% to give an average ascent rate), respectively (Figure 6D; Table 3). The  
751 two Plinian eruptions could be expected to have similar ascent rate however, the 1010 Cal.  
752 CE Plinian eruption is characterised by a fast ascent rate of 0.6 – 12 m/s in comparison the  
753 5680 Cal. BCE Plinian eruption which is characterised by the slowest ascent rate of 0.04 -1  
754 m/s. This difference is related to the feedback loop between ascent rate and microlite  
755 growth which leads to a slower ascent rate in the microlite-rich 5680 Cal. BCE eruption  
756 (Cassidy et al., 2018).

### 757 **5.3 Top-Down, Bottom-Up and External Processes**

758 The transition in explosive eruption style at La Soufrière is not related to any large changes  
759 in the mush storage region or supply system, as shown by the lack of compositional and  
760 volatile differences in the magmas feeding the different eruption styles. The variations in  
761 ascent rate (Fig 6D) indicate that faster ascent rates often lead to more intense eruptions,  
762 the ascent rate variations may relate to mass flux entering the conduit. However, the range  
763 in viscosity changes indicate processes occurring above the magma storage zone and within  
764 the conduit also control eruption style and explosivity. This suggests that external factors  
765 and the interplay of top-down and bottom-up controls must be considered for the La  
766 Soufrière system.

767 At andesitic volcanoes such as La Soufrière, top-down controls are related to the extent and  
768 dynamics of the hydrothermal system on magma in the conduit and the presence of lava  
769 lakes as well as of caldera structures and lava domes (e.g., Endo et al., 1981; Alidibirov and  
770 Dingwell et al., 1996; Sparks, 1997; Scandone et al., 2007; Boudon et al., 2015; Cassidy et al.,  
771 2018; Heap et al., 2019). In this context, fracture-generating processes driven by thermo-  
772 hydraulic circulation, hydrothermal inflation and rock alteration result in the establishment  
773 of connectivity between the lower-pressure upper zones and the deeper pressurised regions  
774 of the plumbing system (Roman and Cashman, 2018). A lava dome can favour pressure  
775 build-up by sealing shallow outgassing fractures (Taisne and Jaupart, 2008; Boudon et al.,  
776 2015; Chevalier et al., 2017) or plugging the conduit (Diller et al., 2006; Collinson and  
777 Neuberg, 2012; Boudon et al., 2015; Bain et al., 2019). Explosions are promoted by smaller

778 domes where pressure build up is sufficient to overcome load pressures; as the lava dome  
779 load increases pressure build up may become insufficient to overcome the lava dome load  
780 pressure and strength (Taisne and Jaupart, 2008; Boudon et al., 2015). Lava domes can also  
781 modulate explosions during long-lived activity. If pressure builds up below a 'plugging' dome  
782 (Johnson et al., 2014), magma may fragment through tensile failure (Hornby et al., 2019)  
783 leading to explosive eruptions. Larger lava domes are also less likely to become efficiently  
784 sealed as deep-seated fractures are more likely to form in the the outer parts of the lava  
785 dome increasing the permeability (Boudon et al., 2015). Sector collapses and phreato-  
786 magmatic interactions may also greatly enhance or even trigger highly explosive activity  
787 (Pinel and Jaupart et al., 2000; Scandone et al., 2007).

788 La Soufrière has a history of dome emplacement, large dome collapse events, high amounts  
789 of external water and also has a well-developed and active hydrothermal system, which  
790 forms quickly at any hot cycle of volcanic activity. Hydrothermal alteration results in  
791 modifications to the shape of the pore network, this can result in reductions in permeability  
792 and porosity, decreasing volcano stability and increasing pore pressure and promoting  
793 explosive behaviour (e.g., Lopez and Williams et al., 1993; Sparks, 1997; Komorowski et al.,  
794 1997; Reid, 2004; Komorowski et al., 2010; Pola et al., 2012; Ball et al., 2013; Wyering et al.,  
795 2015; Navelot et al., 2018; Heap et al., 2019; Heap et al., 2021; Komorowski et al., in prep).  
796 Like for many andesitic volcanoes, these major features of the La Soufrière system must be  
797 considered as controlling effects on magmatic eruption style.

798 The influence of the hydrothermal system alteration at La Soufrière (e.g., Navelot et al.,  
799 2018) and external forcing has already been proposed as important controls on phreatic  
800 eruptions (Zlotnicki et al., 1992; Moune et al., 2022). High fluid circulation combined with  
801 sealing of the dome with pressure unable to be released increases the risk of a phreatic  
802 eruption, as hypothesised for the 1976-77 eruption (Zlotnicki et al., 1992). An alternative  
803 hypothesis proposed for the 1976-77 eruption is the efficient, extensive hydrothermal  
804 system might buffer the rise of small magma batches, leading to their stagnation at shallow  
805 depth and limiting the eruption active to only phreatic explosions (Feuillard et al., 1983;  
806 Villemant et al., 2014). It is therefore also important to understand the potential control of  
807 the hydrothermal system and external forcing on a magma ascent and its eruption potential  
808 and explosivity.

### 809 **5.3.1 The 5680 Cal. BCE Plinian eruption (VEI 4) – slow ascent and the importance of top-** 810 **down controls**

811 As no temperature differences within the error estimates are observed between the  
812 average GM (990°C) and MI (1035°C), this could indicate that any magma moving from the  
813 supply system is not preserved in the erupted area of the storage zone. Instead, processes  
814 of mixing different parts of the same mush may be more relevant (e.g., Solaro et al., 2020).  
815 This could be considered a realistic scenario based on: the lack of GM mixing textures,  
816 homogenous MI composition in both the Type 1, 2, 3 and 6 OPX and Type-4 and 5 OPX and

817 MIs and GM glass show considerable compositional overlap. Alternatively, magmas may  
818 have been well mixed prior to eruption.

819 The final destabilisation of the mush system has affected the majority of the erupted area  
820 of the storage zone (~9 Km depth) 40 days before eruption (Metcalf et al., 2021) (Fig 7A).  
821 In comparison, to the other eruptions discussed here, the 5680 Cal. BCE eruption had a  
822 slow ascent rate (0.04 – 1 m/s).

823 Prior to the 5680 Cal. BCE eruption, lava flow and dome extrusions were identified in the  
824 eruptive record (Samper et al., 2009; Legendre, 2012). This effusive period of activity  
825 followed in eruptive record directly by the explosive Plinian eruption (Legendre, 2012),  
826 with the transition of effusive to explosive activity potentially influenced by the upper  
827 conduit and vent (Sparks, 1997; Platz et al., 2007; Castro and Gardner, 2008; Cassidy et al.,  
828 2018).

829 This transition can be explained by the properties calculated for the 5680 Cal. BCE magma.  
830 The slow ascent observed for this eruption should have allowed volatiles to decouple from  
831 the magma under an open-system degassing regime (Gonnermann and Manga, 2013).  
832 Feedbacks between degassing and crystallization (observed in this eruption which is  
833 characterised by a high microlite content) were able to increase viscosity and slow ascent  
834 further allowing more degassing. Such gas loss would have favoured the occurrence of an  
835 effusive eruption (Melnik and Sparks, 1999; Cassidy et al., 2018) (Fig 7A).

836 However, the syn-eruptive viscosity changes resulting from degassing and crystallization  
837 feedbacks and the conduit sealing related to a lava dome and/or hydrothermal processes  
838 would have reduced outgassing efficiency (Sparks, 1997; Komorowski et al., 1997; 2010).  
839 With the combination of microlite growth creating large pressure gradients and gas  
840 accumulation beneath the sealed dome, leading to pressurisation of the magma (Sparks,  
841 1997; Cassidy et al., 2015).

842 The pressure increase is contained by the elastic deformation of the wall rocks until this  
843 overpressure exceeds the strength of the confining rock and the seal fails resulting in this  
844 case, a Plinian eruption (Cortés et al., 1997, Barmin et al., 2002, Lavallée et al., 2012;  
845 Boudon et al., 2015). This scenario of slow ascending magmas resulting in explosive  
846 eruptions has been shown to occur at several systems including the Inyo volcanic chain,  
847 USA (Castro and Gardner, 2008).

### 848 **5.3.2 The 1010 Cal. CE Plinian phase (VEI 4)– fast ascent and the importance of bottom-** 849 **up controls**

850 Similar to the 5680 Cal. BCE, no temperature differences within the error estimates are  
851 observed between the average groundmass glass (1025°C) and MI (1000°C) for the 1010 Cal.  
852 CE eruption. Again, this could indicate that any magma moving from the supply system did  
853 not directly impact the erupted area of the storage zone. As with 5680 Cal. BCE, mixing  
854 different parts of the same mush may have occurred (e.g., Solaro et al., 2020). This is again  
855 shown by: the lack of GM mixing textures, MIs and GM glass show considerable

856 compositional overlap and homogenous MI composition in the Type 1, 2, 3 and 6 OPX.  
857 Alternatively, magmas may have been well mixed prior to eruption.

858 The final destabilisation of the mush system has affected the majority of the erupted area of  
859 the storage zone (~9 Km depth) 848 days before eruption (Metcalf et al., 2021). In  
860 comparison to the shorter diffusion timescales calculated for other eruptions, which are on  
861 the scale of days to weeks, this indicates that recharge and mixing in the storage zone has  
862 occurred over a long period. Despite the magma's long residence in the storage zone,  
863 magma has ascended relatively rapidly in the conduit (0.6 -12 m/s) in comparison to the  
864 other eruptions.

865 The 1010 Cal. CE explosive eruption was followed by an effusive phase (Legendre, 2012).  
866 Though the previous eruption (450 Cal. CE) was an effusive eruption, it is unclear whether  
867 prior to the 1010 Cal. CE eruption a dome was emplaced, if a dome was present our results  
868 suggest that the state of the upper conduit and vent has not had a major control on the  
869 1010 Cal. CE eruption (Fig 7B).

870 Instead, volatile exsolution drove rapid ascent by increasing buoyancy, during fast ascent  
871 outgassing is less efficient with exsolved volatiles remain coupled to the melt in a closed-  
872 system degassing scenario (Mangan and Sisson, 2000; Cashman, 2004; Szramek et al., 2006).  
873 The small syn-eruptive viscosity change observed for the 1010 Cal. CE and microlite-poor  
874 groundmass indicates that decompression-induced crystallisation was limited, and has  
875 resulted in fluid-like behaviour by reducing viscosity and further increasing magma  
876 buoyancy (Anderson, 1995; Martel, 1998; Scandone et al., 2007; Massol and Jaupart, 1999;  
877 Martel, 2012). This rapid ascent of a magma with volatiles coupled to the melt in a closed  
878 system led to high overpressures and promoted explosive behaviour. Driven from the  
879 'bottom-up' the rapid ascent of a gas-rich magma led to explosive fragmentation and a  
880 Plinian eruption (Cassidy et al., 2018; Fig 7B). A slower ascent rate, as in the case of 1657  
881 Cal. CE may have resulted in a less explosive eruption (e.g., Szramek et al., 2006).

### 882 **5.3.3 The 1530 Cal. CE Sub-Plinian phase (VEI 3) – the importance of external forces**

883 The 1530 Cal. CE eruption began with injection of a less evolved magma from the supply  
884 system into the storage system (Pichavant et al., 2018). Magma then ascended at 2-4 m/s  
885 through the conduit, an ascent rate that is most comparable to the magma ascent rate for  
886 the 1657 Cal. CE eruption.

887 The microlite-poor (17 vol%) glass observed in the light bands of the banded 1530 Cal. CE  
888 eruption products, are comparable to the 1657 Cal. CE groundmass glasses (18 vol%).  
889 However, the 1530 Cal. CE has the additional homogenous glass (27 vol%) and glass from  
890 dark bands (34 vol%) that are compositionally different. This wider range of textures  
891 indicates more complex processes affected the conduit.

892 The 1530 Cal. CE eruption is the most well-constrained La Soufrière eruption. It began with a  
893 phreatic phase followed by a flank collapse which preceded the arrival of magma at the  
894 surface and fed the sub-Plinian explosive phase discussed throughout this paper



895 (Komorowski et al., 2005; 2008; Touboul et al., 2007; Boudon et al., 2008; Pichavant et al.,  
896 2018). The andesite magma was erupted first transitioning through banded black and white  
897 pumice and then to basaltic-andesite. Following the sub-Plinian phase the eruption style  
898 changed to strombolian activity and a less explosive phase, with the end of the multiphase  
899 eruption marked by the growth of the current andesite lava dome (Boudon et al., 2008).

900 The flank collapse prior to the sub-Plinian phase indicates that external forces have had a  
901 major control on the generation of this magmatic eruption (Boudon et al., 2003; Boudon et  
902 al., 2007). The volcanic edifice can exert a large load which affects the stress field at depth  
903 reservoir (Pinel and Jaupart, 2000, 2005). A flank collapse rapidly releases this load resulting  
904 in a downward propagating pressure change which affects the decompression rate of the  
905 magma resulting in fragmentation and in this case, contribute to the triggering of a sub-  
906 Plinian eruption style (Boudon et al., 2003; Pinel and Jaupart, 2005; Gudmundsson, 2006;  
907 Scandone et al., 2007).

908 The stress fields exerted by the edifice can also affect the petrological and chemical  
909 evolution of the magma in the storage zone and can prevent denser, less evolved magmas  
910 reaching the surface (Pinel and Jaupart, 2000, 2005; Gudmundsson, 2006). At several  
911 volcanoes including. Mount St. Helens (Pinel and Jaupart, 2000), Bezymianny volcano  
912 (Kamchatka, Russia) (Izbekov et al., 2006) and volcanoes in the Lesser Antilles including on  
913 Martinique and St Lucia (Boudon et al., 2013), the unloading of the volcanic edifice and  
914 change in stress field from events such as flank collapse have been shown to result in a  
915 change in magma composition to more mafic compositions erupted after the collapse event  
916 (Pinel and Jaupart, 2000). This may be a plausible scenario for the 1530 Cal. CE eruption  
917 with changes in the stress field controlling the transition from andesite to black and white  
918 banded pumice and then to basaltic-andesite which were present in the storage zone  
919 following the recharge event. In comparison, the other eruptions do not have a banded  
920 component and flank collapses were not thought to have occurred directly before a  
921 magmatic explosive eruption.

#### 922 **5.3.4 The 1657 Cal. CE Vulcanian eruption (VEI 2) – fast ascent and the importance of** 923 **bottom-up controls**

924 As with the two Plinian eruptions no temperature differences within the error estimates are  
925 observed between the average groundmass glass ((975°C) and MI (1000°C) for the 1010 Cal.  
926 CE eruption. There are also no GM mixing textures, MIs and GM glass show considerable  
927 compositional overlap and homogenous MI composition in the Type 1, 2, 3 and 6 OPX.  
928 There is also some bimodal variation in the groundmass glass compositions. This suggests  
929 mafic recharge may have occurred prior to this eruption and resulted in incomplete mixing.  
930 The final destabilisation of the mush system has affected the majority of the erupted area of  
931 the storage zone 35 days before eruption (Metcalf et al., 2021). Eruptible magma then  
932 ascended from ~8 Km depth in the storage region into the conduit and then to the surface.

933 Though this eruption did not have an effusive phase, the 1530 Cal. CE dome, still present  
934 today, was emplaced prior to the 1657 Cal. CE eruption (Komorowski et al., 2005; Boudon et  
935 al., 2008; Legendre, 2012). For the 5680 cal. BCE eruption the dome and top-down controls  
936 have had a major effect on producing the explosive eruption. However, for 1657 Cal. CE the  
937 evidence for the occurrence of similar major top-down controls is not clear.

938 Given the composition and volatile contents of the 1010 Cal. CE and 1657 Cal. CE eruptions  
939 are very similar, the magma could have been expected to ascend in similar ways and to lead  
940 similar explosive eruptions. However, magma in the 1657 Cal. CE eruption had a slower  
941 ascent rate and experienced a larger syn-eruptive viscosity change. The slower ascent rate  
942 may have allowed more time for microlite crystallisation in the magma of the 1657 Cal. CE  
943 eruption than in the 1010 Cal. CE eruption, a process that in turn increased its viscosity  
944 (Sparks, 1997; Melnik and Sparks, 1999). The viscosity change that resulted from microlite  
945 crystallisation in a closed-system promoted some limited explosivity of the small magma  
946 batch that only led to a Vulcanian eruption (Fig 7D). If the ascent rate had been faster, as  
947 with the 1010 Cal. CE eruption, this may have resulted in a more explosive eruption. The  
948 absence of dome collapse during this small eruption suggests that pressure in the magma  
949 column overcome the load of the dome to produce a small explosive eruption (Fig 7D).

### 950 **5.3.5 Implications for future eruptions and monitoring**

951 This data suggests shows that explosive eruption style at La Soufrière is controlled by a  
952 combination of ascent rate and top-down controls and not necessarily linked exclusively to  
953 bottom-up processes, as highlighted by the Plinian eruptions studied here. Looking towards  
954 a future eruption, the various scenarios leading to explosive magmatic eruptions, and  
955 particularly the multiple controls on Plinian eruptions, make the La Soufrière volcanic  
956 system more hazardous and complex to forecast.

957 The results of this study have wider implications, not just for La Soufrière but for other  
958 systems with a developed hydrothermal system that tends to promote sealing of the host-  
959 rock, which influences the dynamics of magma ascent and degassing at shallow depth (e.g.,  
960 Komorowski et al., 1997; 2010; in prep). To improve forecasting of potential future  
961 magmatic eruptions, continued development of volcano monitoring techniques must focus  
962 on developing the ability to: 1) reconstruct, via real-time geodesy, the deep stress field  
963 changes behind the upward movement of magma; 2) track the modifications of the physical  
964 properties of the host-rock; and 3) the development and evolution of sealing in the  
965 hydrothermal system. To understand the evolution of the magmatic system and how a  
966 potential eruption could evolve, monitoring should also keep tracking the petrology and  
967 rock properties of early erupted products, imaging the physio-chemical properties of the  
968 host-rock and its structures in time and space, and quantifying external forcing processes of  
969 the hydrothermal system that can modulate magma ascent dynamics. Particularly for  
970 volcanic systems with a wide range of eruptive behaviour, such as La Soufrière, this could  
971 provide a more complete understanding of how an eruption could develop and what its  
972 credible expected intensity might be.

## 973 **6 Conclusions**

974 The variation in eruption style at La Soufrière gives us opportunity to understand the  
975 controls on eruption explosivity at this volcano. The four eruptions discussed here are a  
976 Vulcanian (VEI 2), sub-Plinian (VEI 3) and two Plinian eruptions (VEI 4), covering a range of  
977 dominantly explosive magmatic activity produced by La Soufrière. Major and volatile  
978 elements of melt inclusions across the eruption series show the composition of the mush  
979 storage zone has remained stable. Melt inclusions also allow us to constrain the mush  
980 storage zone to 7 – 9 Km depth with an average temperature of 1000°C. The constant  
981 composition and physical properties of the storage zone feeding the eruptions indicate that  
982 for some eruptions processes occurring from the top-down and external forces have also  
983 determined the magma ascent rate and controlled eruption style, particularly the intensity  
984 of the explosive activity.

985 Magma feeding the 1010 Cal. CE eruption ascended relatively rapidly (0.6 – 12 m/s)  
986 preventing the growth of microlites. The magma and exsolved gas remain coupled and  
987 ascended rapidly to trigger an explosive eruption style that was primarily driven by bottom-  
988 up processes. In contrast to the 1010 Cal. CE eruption, the 1657 Cal. CE eruption, where the  
989 magma ascended at a slower ascent rate (0.7 – 3 m/s), we find little geochemical and  
990 petrological evidence for any major top-down controls. This suggests the differences in  
991 ascent rate may have resulted in contrasting explosive eruption styles.

992 The magma feeding the 5680 Cal. BCE ascended comparatively slowly (0.04 -1 m/s) due to  
993 feedbacks between degassing and crystallization. These processes lead to an increase  
994 viscosity, further slowing magma ascent and promoting magma degassing. In the absence of  
995 any top-down controls this magma could have been expected to erupt with a low explosivity  
996 or effusively. However, sealing resulting from the capping dome and a reduction in  
997 permeability in the conduit favoured the accumulation of gas, leading to over pressurisation  
998 and an explosive Plinian eruption. External forces also played a role in the 1530 Cal. CE  
999 eruption where a flank collapse has resulted in rapid decompression of a gas-rich magma at  
1000 shallow depth that rapidly led to a phase of sub-Plinian explosive activity at the onset of the  
1001 multiphase eruption.

1002 The multiple controls on explosive eruption style, and particularly on the most intense  
1003 eruptions observed at La Soufrière, Plinian (VEI 4) eruptions, render this volcanic system  
1004 particularly hazardous. Hence, the forecasting of the most credible eruption scenario is  
1005 particularly challenging. Given that several eruption styles are possible and that eruption  
1006 style can evolve within an eruption, we stress that monitoring efforts must also address the  
1007 issue of tracking ascent and shallow-depth conduit processes which can influence eruption  
1008 style, and particularly the intensity of explosivity. Given these uncertainties, we recommend  
1009 that an integrated risk-reduction approach consider the capacity to detect rapid changes in  
1010 unrest dynamics and associated eruption-driving processes, both at depth but also near the  
1011 surface, and to develop associated preventive crisis response strategies on a short time  
1012 frame.

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## 1042 **8 Author Contributions**

1043 AM prepared the samples, acquired the geochemical and petrological data, processed and  
1044 analyzed all the data, wrote the draft of the manuscript and drafted the figures. J-CK, SM,  
1045 and AM undertook fieldwork, and sampling. All authors discussed the data, wrote and  
1046 revised the manuscript and the figures.

1047

## 1048 **9 Figure Captions**

1049 **Figure 1:** Map of southern Basse-Terre showing the location of La Soufrière and volcanic  
1050 complexes which make up the island. Also shown are sampling locations for each eruption.

1051 Inset Left: Map showing the position of Guadeloupe in the Lesser Antilles Island arc chain.  
1052 Inset Right: Image of La Soufrière (2020, Metcalfe).

1053

1054 **Figure 2:** Example images of glass from La Soufrière. **(A)** Plagioclase and **(B)** pyroxene hosted  
1055 melt inclusions. Only inclusions with no daughter crystals analysed, inclusions can be round  
1056 and uniform or more irregular. Plagioclase often shows primary and secondary inclusions,  
1057 only inclusions from the core of the crystal which are primary inclusions were analysed. **(C-**  
1058 **E)** Pyroxene hosted inclusions (C – 5680 Cal. BCE, D – 1010 Cal CE, E – 1657 Cal CE) **(F-H)**  
1059 Plagioclase hosted inclusions (F – 5680 Cal. BCE, G – 1010 Cal CE, H – 1657 Cal CE). **(I-N)**  
1060 Groundmass glass (I – 5680 Cal. BCE, J – 1010 Cal CE (Legendre, 2012), K – 1657 Cal CE  
1061 (Legendre, 2012), L – 1530 Cal CE (light glass) (Martel et al., 2021), M – 1530 Cal CE (dark  
1062 glass) (Martel et al., 2021), N - 1530 Cal CE (homogenous glass) (Martel et al., 2021).

1063

1064 **Figure 3:** Total Alkali-Silica whole rock dataset for the La Soufrière eruptions and parental  
1065 Monts Caraïbes magma. The groundmass glass show some Na<sub>2</sub>O loss during analysis.  
1066 Generally, the data falls on the same liquid-line of descent.

1067

1068 **Figure 4:** Complete major element data set: (A) FeO, (B) CaO, (C) K<sub>2</sub>O, (D) MnO, (E) MgO, (F)  
1069 Al<sub>2</sub>O<sub>3</sub>, (G) Na<sub>2</sub>O, (H) TiO<sub>2</sub> vs SiO<sub>2</sub>.

1070

1071 **Figure 5:** Volatile contents for melt inclusion and: (A) H<sub>2</sub>O, (B) CO<sub>2</sub>, (C) S, (D) Cl, (E) F vs K<sub>2</sub>O  
1072 and (F) Total Volatile Content vs VEI. Data is clustered with no significant difference in the  
1073 volatile contents for each eruption.

1074

1075 **Figure 6:** Eruption year vs (A) Temperature (Putirka, 2008), (B) Pressure (Papale et al., 2006,  
1076 errors calculated based on model error and volatile analysis error), (C) Viscosity  
1077 (groundmass (GM) and groundmass with microlites (GM+X), Mastin, 2002), (D) Ascent Rate  
1078 (Mastin, 2002).

1079

1080 **Figure 7:** Schematic diagram of the La Soufrière reservoir for each eruption discussed, with  
1081 calculated parameters annotated. **(A)** 5680 Cal. BCE (VEI 4): eruptible melt from 9 km depth  
1082 begins ascending in the conduit at max. 1 m/s. The slow ascent results in feedbacks  
1083 between degassing and crystallization increasing viscosity and slowing ascent further.  
1084 Sealing of the conduit from the top-down due to the presence of a dome results in exsolved  
1085 volatile accumulation and pressure increases, until the overpressure exceeds the strength of  
1086 the confining rock and the seal fails resulting in a Plinian eruption. **(B)** 1010 Cal. CE (VEI 4):  
1087 eruptible melt from 6 Km depth begins ascending in the conduit at max. 12 m/s. Volatile  
1088 exsolution has prevented a large viscosity change but increased buoyancy allowing the  
1089 magma to rise. Disequilibrium closed system degassing has driven magma still coupled to  
1090 the melt from the bottom up allowing fast ascent resulting in a Plinian eruption. **(C)** 1530  
1091 Cal. CE (VEI 3): magma from the supply system moves up into the storage zone resulting in

1092 magma mixing. Eruptible melt from 7 Km depth begins ascending in the conduit at 2-4 m/s,  
1093 with microlite textures reveal a complex ascent history. A flank collapse has rapidly released  
1094 the edifice load resulting in a downward propagating pressure change which affects the  
1095 decompression rate of the magma resulting in fragmentation a sub-Plinian explosion. **(D)**  
1096 1657 Cal. CE (VEI 2): eruptible melt from 8 Km depth begins ascending in the conduit at max  
1097 3 m/s, the slower ascent compared to the 1010 CE eruption and without influence from top  
1098 down processes, has resulted in a Vulcanian explosion. The dome did not collapse during  
1099 this small eruption.

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1610 **Table 1:** Summary of groundmass glass and microlites for each eruption.

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1620 **Table 2:** Full normalised MI dataset including major elements from EMPA and volatile data from EMPA and SIMS (\*). Also shown are the enstatite (En) and  
1621 anorthite (An) content of the host mineral and the calculated Kd value. Some volatiles were less than the detection limit (<dl).

Table 1							
Eruption	Style	VEI	Glass	Microlites	Microlite size (µm)	Vesicles	Microlite vol%
1657 Cal. CE	Vulcanian	2	Homogenous	subhedral - euhedral	<10	yes	18
1530 Cal. CE	Sub-Plinian	3	Heterogeneous	subhedral - euhedral	<50	yes	17-34
1010 Cal. CE	Plinian	4	Homogenous	anhedral	<5	yes	8
5680 Cal. BCE	Plinian	4	Homogenous	anhedral - subhedral	1 - 15	yes	47

Table 2																			
Eruption	Sample	Host Crystal En	Host Crystal An	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	F	S	Cl	H <sub>2</sub> O	CO <sub>2</sub>	δD	Kd
1657 CE	0913D-pyx7-1	0.39	-	77.74	0.38	12.19	2.50	0.02	0.47	2.05	2.12	2.53	<dl	0.037	0.16	-	-	-	0.23
1657 CE	0913D-pyx7-2	0.39	-	76.72	0.39	12.24	3.14	0.12	0.54	2.38	2.40	2.07	<dl	0.019	0.18	-	-	-	0.24
1657 CE	0913D-pyx7-3	0.39	-	77.01	0.42	12.46	2.94	0.08	0.49	2.45	2.27	1.87	<dl	0.028	0.19	-	-	-	0.22
1657 CE	0913D-pyx-9-1	0.57	-	75.30	0.47	12.70	3.44	0.18	0.65	2.71	2.74	1.80	<dl	0.056	0.25	-	-	-	0.24
1657 CE	0913D-pyx16-1	0.39	-	74.79	0.43	13.58	2.76	0.03	0.86	3.33	2.45	1.76	0.051*	0.006*	0.29*	3.6*	530*	-10.1	0.28
1657 CE	0913D-pyx16-2	0.38	-	75.23	0.45	13.59	2.91	0.09	0.45	3.02	2.50	1.77	0.050*	0.005*	0.24*	3.7*	674*	-25.6	0.24
1657 CE	0913D-pyx-16-3	0.39	-	75.26	0.43	13.57	3.05	0.12	0.53	3.22	2.32	1.49	0.048*	0.005*	0.23*	3.6*	451*	-10.9	0.24
1657 CE	0913D-plg7-1	-	0.68	76.33	0.50	13.97	2.15	0.11	0.28	2.73	1.84	2.09	0.048*	0.005*	0.24*	3.4*	146*	-10.5	0.08
1657 CE	0913D-plg7-2	-	0.66	76.94	0.45	13.56	2.33	0.11	0.39	2.72	1.57	1.94	0.049*	0.006*	0.25*	3.9*	165*	-46.1	0.10
1657 CE	0913D-plg11-2	-	0.68	74.97	0.75	14.23	2.14	0.10	0.33	3.10	2.71	1.67	0.039*	0.006*	0.21*	2.4*	35*	-40.3	0.07
1657 CE	0913D-plg11-3	-	0.70	74.22	0.60	13.80	1.99	0.08	0.35	3.44	3.41	2.12	0.045*	0.007*	0.32*	3.7*	122*	-37.1	0.05

1657 CE	0913D-plg5-2	-	0.69	73.13	0.53	12.50	4.67	0.14	0.87	3.06	2.97	2.11	0.046*	0.009*	0.36*	3.9*	192*	-26.6	0.05
1657 CE	0913D-plg2-1	-	0.75	75.90	0.45	13.90	2.05	0.01	0.31	2.73	3.29	1.37	0.04*	0.003*	0.19*	2.3*	253*	-43.6	0.06
1657 CE	0913D-plg3-1	-	0.29	74.49	0.57	13.13	3.88	0.08	0.79	2.60	2.33	2.13	<dl	<dl	0.32	-	-	-	0.06
1657 CE	0913D-plg3-2	-	0.97	74.98	0.55	12.70	4.28	0.15	0.76	2.85	1.68	2.04	<dl	0.021	0.33	-	-	-	0.07
1657 CE	0913D-plg3-3	-	0.97	74.82	0.60	12.51	3.78	0.13	0.76	2.96	2.22	2.24	<dl	0.015	0.30	-	-	-	0.07
1657 CE	0913D-plg3-4	-	0.57	74.71	0.64	12.47	3.77	0.12	0.77	2.74	2.53	2.24	<dl	0.025	0.32	-	-	-	0.05
1657 CE	0913D-plg1-1	-	0.58	75.69	0.56	12.55	3.29	0.04	0.58	2.63	2.53	2.13	<dl	0.007*	0.29*	2.4*	35*	-39.4	0.08
1657 CE	0913D-plg1-2	-	0.76	75.37	0.58	12.49	3.37	0.07	0.59	2.72	2.62	2.19	<dl	0.036	0.29	-	-	-	0.05
1657 CE	0913D-plg5-1	-	0.66	74.94	0.53	13.42	4.69	0.16	0.80	3.04	0.82	1.59	0.05*	0.009*	0.36*	4.1*	36*	-50.8	0.05
1657 CE	0913D-plg1-3	-	0.69	74.66	0.46	13.07	3.29	0.13	0.56	3.06	2.64	2.13	<dl	0.021	0.272	-	-	-	0.06
1657 CE	0913D-plg5A-1	-	0.59	76.20	0.31	13.11	2.44	0.01	0.34	2.75	2.59	2.25	<dl	0.010	0.221	-	-	-	0.07
1657 CE	0913D-plg5A-2	-	0.61	75.94	0.34	13.51	2.28	0.10	0.32	3.00	2.41	2.10	<dl	0.042	0.089	-	-	-	0.09
1657 CE	0913D-plg8-1	-	0.57	77.61	0.48	12.36	2.38	0.02	0.39	2.28	2.31	2.17	<dl	<dl	0.225	-	-	-	0.07
1657 CE	0913D-plg8-2	-	0.58	78.16	0.55	12.43	1.78	0.10	0.39	2.11	2.36	2.12	<dl	0.022	0.211	-	-	-	0.07
1657 CE	0913D-plg4-1	-	0.59	77.12	0.59	12.34	2.44	0.12	0.42	2.47	2.47	2.03	<dl	<dl	0.208	-	-	-	0.07
1657 CE	0913D-plg4-2	-	0.59	77.31	0.59	12.25	2.42	0.17	0.35	2.27	2.54	2.09	<dl	0.017	0.196	-	-	-	0.06
1657 CE	0913D-plg4-3	-	0.59	77.75	0.51	12.34	2.16	0.04	0.33	2.27	2.61	1.99	<dl	0.010	0.187	-	-	-	0.06
1657 CE	0913D-plg4-4	-	0.59	77.46	0.53	12.25	2.48	0.13	0.34	2.31	2.49	2.02	<dl	<dl	0.201	-	-	-	0.06
1657 CE	0913D-plg4-5	-	0.59	77.77	0.53	12.36	2.24	0.03	0.35	2.49	2.17	2.06	<dl	<dl	0.201	-	-	-	0.08
1657 CE	0913D-plg4-6	-	0.59	77.65	0.53	12.20	2.25	0.10	0.34	2.33	2.51	2.10	<dl	0.017	0.199	-	-	-	0.05
1657 CE	0913D-plg6-1	-	0.64	77.28	0.50	12.85	2.05	0.02	0.27	2.25	2.43	2.35	<dl	<dl	0.194	-	-	-	0.05
1657 CE	0913D-plg6-2	-	0.67	76.37	0.51	13.24	2.07	0.05	0.28	2.69	2.61	2.17	<dl	0.026	0.182	-	-	-	0.06
1010 CE	0924A-pyx1A-3	0.56	-	69.50	0.67	14.77	5.36	0.21	0.99	3.75	3.13	1.62	<dl	<dl	0.20	-	-	-	0.24
1010 CE	0924A-pyx1D-1	0.56	-	69.87	0.66	14.53	5.03	0.18	0.93	3.90	3.25	1.65	<dl	0.065	0.20	-	-	-	0.24
1010 CE	0924A-pyx1D-2	0.56	-	70.33	0.64	14.46	5.07	0.14	0.89	3.53	3.27	1.69	<dl	0.022	0.21	-	-	-	0.23
1010 CE	0924A-pyx1D-4	0.59	-	70.66	0.61	14.49	4.57	0.10	0.92	3.62	3.20	1.83	0.035*	0.006*	0.2*	2.2*	11*	-29.5	0.23
1010 CE	0924A-pyx1G-1	0.57	-	69.66	0.67	14.79	4.98	0.14	0.91	3.98	3.37	1.50	<dl	0.046	0.19	-	-	-	0.23
1010 CE	0924A-pyx1G-4	0.57	-	70.30	0.45	14.51	5.12	0.08	0.93	4.00	3.18	1.43	<dl	0.012	0.19	-	-	-	0.23
1010 CE	0924A-pyx2C-1	0.57	-	69.51	0.57	14.69	5.28	0.12	1.03	3.81	3.46	1.53	<dl	0.042	0.21	-	-	-	0.24
1010 CE	0924A-pyx2C-2	0.56	-	70.41	0.52	14.36	4.84	0.09	0.88	3.87	3.47	1.56	<dl	0.031	0.19	-	-	-	0.22
1010 CE	0924A-pyx1B-1	0.41	-	70.80	0.55	14.50	4.86	0.23	0.85	3.80	2.87	1.55	<dl	0.015	0.21	-	-	-	0.25
1010 CE	0924A-pyx13-2	0.60	-	73.28	0.65	14.36	4.90	0.10	1.01	3.19	1.08	1.44	0.041*	0.006*	0.16*	<dl	<dl	-	0.23

1010 CE	0111A-pyx10-2	0.57	-	73.85	0.53	15.43	3.67	0.07	0.68	3.92	0.51	1.33	0.043*	0.01*	0.18*	3.3*	60*	-34.5	0.22
1010 CE	0924A-pyx2F-1	0.54	-	64.32	0.31	21.31	2.34	0.04	0.38	7.04	3.16	1.11	<dl	0.006	<dl	-	-	-	0.25
1010 CE	0111A-pyx10-1	0.57	-	73.44	0.77	15.74	3.63	0.14	0.28	4.04	0.80	1.15	0.048*	0.02	0.24*	4.4*	122*	-30.5	0.22
1010 CE	0924A-pyx6-1	0.61	-	70.28	0.58	15.35	5.28	0.12	0.92	3.47	2.76	1.24	0.042*	0.01*	0.15*	2.8*	131*	-34.5	0.23
1010 CE	0924A-plg2E-1	-	0.67	73.25	0.57	13.92	4.04	0.16	0.78	3.18	2.25	1.85	<dl	0.020	0.17	-	-	-	0.09
1010 CE	0924A-plg2E-2	-	0.67	72.50	0.44	14.12	3.55	0.10	0.77	3.48	3.26	1.77	<dl	0.023	0.16	-	-	-	0.07
1010 CE	0924A-plg2E-3	-	0.60	72.68	0.54	14.04	3.85	0.05	0.83	3.25	2.85	1.92	<dl	0.031	0.18	-	-	-	0.10
1010 CE	0924A-plg2B-1	-	0.62	75.52	0.49	12.41	2.90	0.06	0.48	2.51	3.36	2.26	<dl	0.039	0.32	-	-	-	0.05
1010 CE	0924A-plg2B-2	-	0.61	75.08	0.63	12.57	2.86	0.00	0.42	2.69	3.35	2.39	<dl	0.025	0.29	-	-	-	0.06
1010 CE	0924A-plg2A-1	-	0.61	77.47	0.37	12.28	2.54	0.08	0.34	2.22	2.30	2.39	<dl	0.010	0.25	-	-	-	0.06
5680 BCE	1101viA-pyx2E-2	0.64	-	64.62	0.79	16.08	8.19	0.17	2.75	6.43	0.24	0.73	<dl	0.08	0.17	-	-	-	0.30
5680 BCE	1101viA-pyx2E-3	0.65	-	66.18	0.72	14.76	7.16	0.12	2.50	4.35	3.09	1.12	<dl	0.02	0.21	-	-	-	0.30
5680 BCE	1101viA-pyx2E-4	0.62	-	66.74	0.61	14.79	6.94	0.16	2.54	4.53	2.50	1.18	<dl	0.04	0.20	-	-	-	0.36
5680 BCE	1101viA-pyx2F-3	0.55	-	76.49	0.55	13.34	4.09	0.08	0.87	2.99	0.09	1.49	<dl	<dl	0.22	-	-	-	0.29
5680 BCE	1101viA-pyx1-1	0.58	-	69.38	0.44	13.77	8.03	0.28	1.00	4.24	1.56	1.32	0.048*	0.02*	0.23*	4.2*	866*	-59	0.26
5680 BCE	1101viA-pyx1-2	0.56	-	69.54	0.47	14.76	6.62	0.31	0.81	4.16	1.88	1.46	0.044*	0.02*	0.19*	3.5*	448*	-33.1	0.25
5680 BCE	1101viA-pyx2A-2	0.37	-	76.41	0.49	12.77	4.25	0.17	1.13	3.50	0.05	1.24	<dl	0.02	0.26	-	-	-	0.31
5680 BCE	1101viA-pyx2A-1	0.34	-	75.01	0.55	12.38	3.88	0.13	0.87	3.40	1.62	2.17	<dl	0.02	0.13	-	-	-	0.25
5680 BCE	1101viA-pyx2B-3	0.37	-	74.93	0.59	13.46	4.00	0.10	0.91	3.45	0.92	1.63	<dl	0.05	0.23	-	-	-	0.26
5680 BCE	1101viA-pyx2E-1	0.64	-	64.88	0.64	15.40	7.04	0.20	2.84	4.71	312	1.17	<dl	0.03	0.23	-	-	-	0.36
5680 BCE	1101viA-pyx5-1	0.62	-	68.67	0.82	17.01	3.49	0.22	0.63	4.74	3.03	1.38	0.043*	0.02*	0.19*	3.4*	129*	-64.8	0.35
5680 BCE	1101viA-pyx5-2	0.59	-	64.68	1.00	17.54	7.55	0.15	1.22	6.65	008	1.13	0.051*	0.02*	0.2*	3.1*	182*	-50.8	0.23
5680 BCE	1101viA-plg1D-1	-	0.69	76.13	0.34	14.17	3.02	0.12	0.38	2.75	1.22	1.87	0.06*	0.003*	0.14*	2.4*	116*	-33.2	0.12
5680 BCE	1101viA-plg1D-2	-	0.73	75.61	0.49	12.85	2.59	0.14	0.47	3.02	2.78	2.06	<dl	0.01	0.24	-	-	-	0.04
5680 BCE	1101viA-plg1D-3	-	0.67	76.68	0.51	12.93	2.83	0.07	0.48	2.79	1.66	2.04	<dl	0.02	0.23	-	-	-	0.09
5680 BCE	1101viA-plg1C-3	-	0.77	76.39	0.50	13.41	2.91	0.06	0.43	2.86	1.46	1.99	<dl	0.01	0.25	-	-	-	0.06
5680 BCE	1101viA-plg1C-3	-	0.68	76.63	0.37	13.36	2.82	0.07	0.40	2.84	1.46	2.04	<dl	0.01	0.21	-	-	-	0.10
5680 BCE	1101viA-plg1A-1	-	0.01	75.68	0.64	12.84	3.14	0.23	0.55	2.86	2.09	1.98	<dl	0.02	0.22	-	-	-	0.07
5680 BCE	1101viA-plg1A-2	-	0.68	76.27	0.70	12.73	3.36	0.01	0.56	2.87	1.62	1.88	<dl	0.02	0.24	-	-	-	0.09
5680 BCE	1101viA-plg1E-2	-	0.70	74.97	0.42	13.72	2.75	0.02	0.43	3.36	2.41	1.90	<dl	<dl	0.21	-	-	-	0.07
5680 BCE	1101viA-plg1B-1	-	0.67	76.25	0.48	13.12	3.27	0.09	0.46	2.87	1.57	1.90	<dl	0.01	0.18	-	-	-	0.10
5680 BCE	1101viA-plg1B-2	-	0.66	76.33	0.47	12.93	3.60	0.04	0.47	2.98	141	1.77	<dl	<dl	0.10	-	-	-	0.12

5680 BCE	1101viA-plg1B-3	-	0.71	73.31	0.57	12.28	3.73	0.03	1.42	3.88	2.89	1.90	<dl	0.02	0.13	-	-	-	0.06
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1622

1623 **Table 3:** Summary table of the analysed data and calculated properties (including temperature for MI and GM, pressure and viscosity (GM without

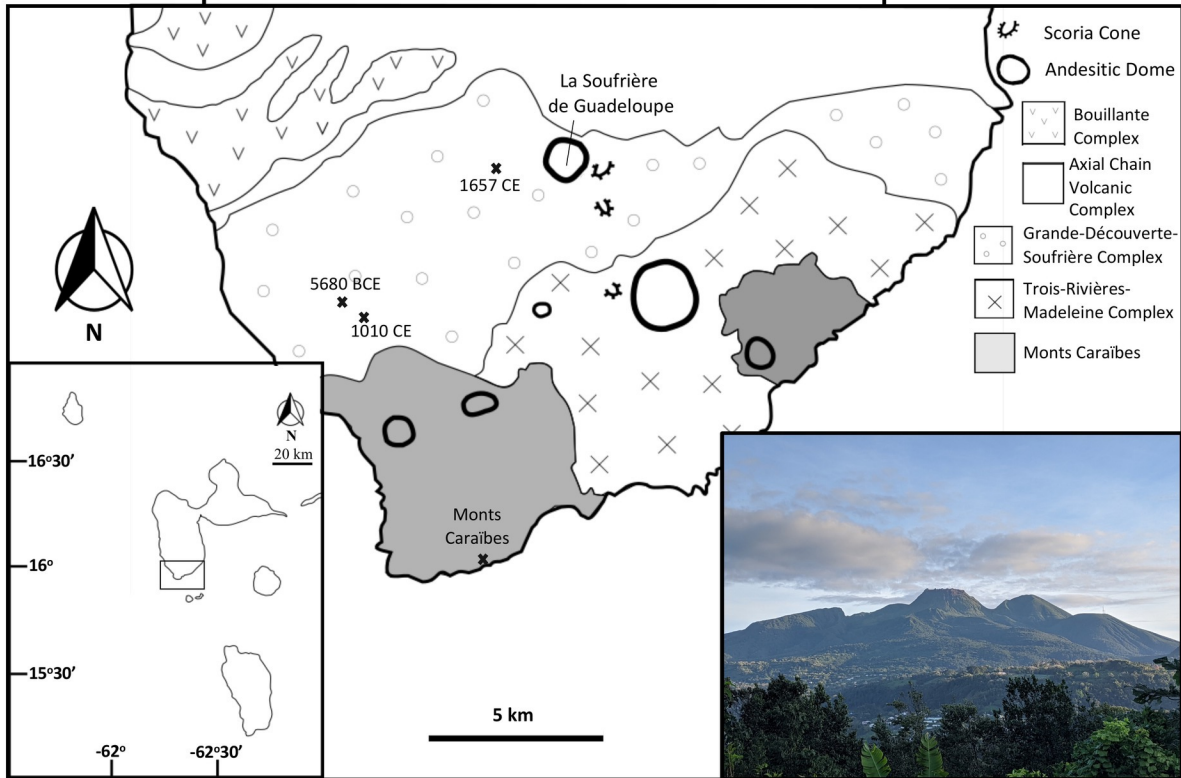
1624 microlites and GM+X with microlites) \*Boudon et al., 2008; Pichavant et al., 2018; Poussineau, 2005

Eruption	Eruption Style	VEI	Measured Properties					Calculated Properties						
			Av. Bulk Rock SiO <sub>2</sub> (wt%)	Av. MI SiO <sub>2</sub> (wt%)	Max. H <sub>2</sub> O (wt%)	Max. CO <sub>2</sub> (ppm)	Microlites (vol%)	Av. MI Temp (°C)	Av. GM Temp (°C)	Pressure (MPa)	Depth (Km)	Viscosity (Pa·s)		Ascent Rate (m/s)
												GM	GM+X	
1657 Cal. CE	Vulcanian	2-3	58.1	76.1	4.14	674	18	1000	975	190	7.7	10 <sup>7.6</sup>	10 <sup>8</sup>	0.7 - 3
1530 Cal. CE	Sub-Plinian	3	57.5*	74.3*	5*	-	17 - 34	875*	1000*	170*	7*	10 <sup>8.2</sup>	10 <sup>8.5</sup>	2 - 4
1010 Cal. CE	Plinian	4	58	71.6	4.42	131	8	1000	1025	210	8.6	10 <sup>6</sup>	10 <sup>6</sup>	0.6 - 12
5680 Cal. BCE	Plinian	4	-	72.7	4.19	866	47	1035	990	220	8.7	10 <sup>5.4</sup>	10 <sup>7</sup>	0.04 - 1

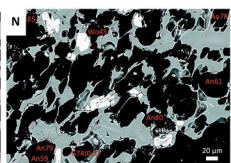
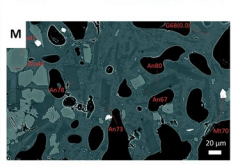
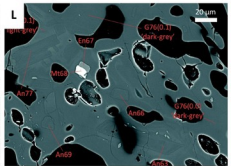
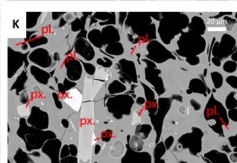
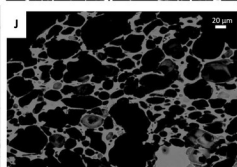
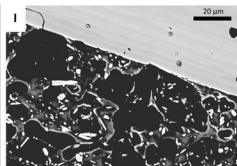
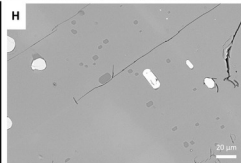
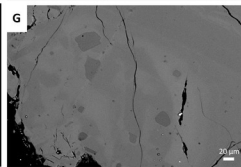
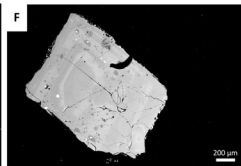
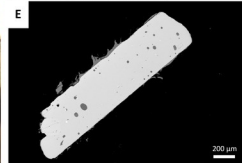
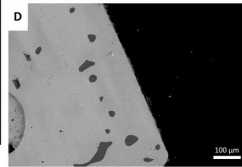
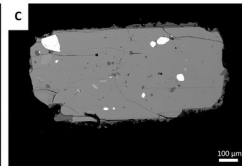
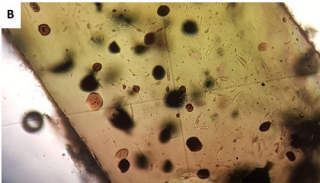
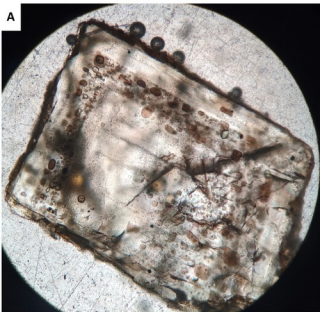
1625

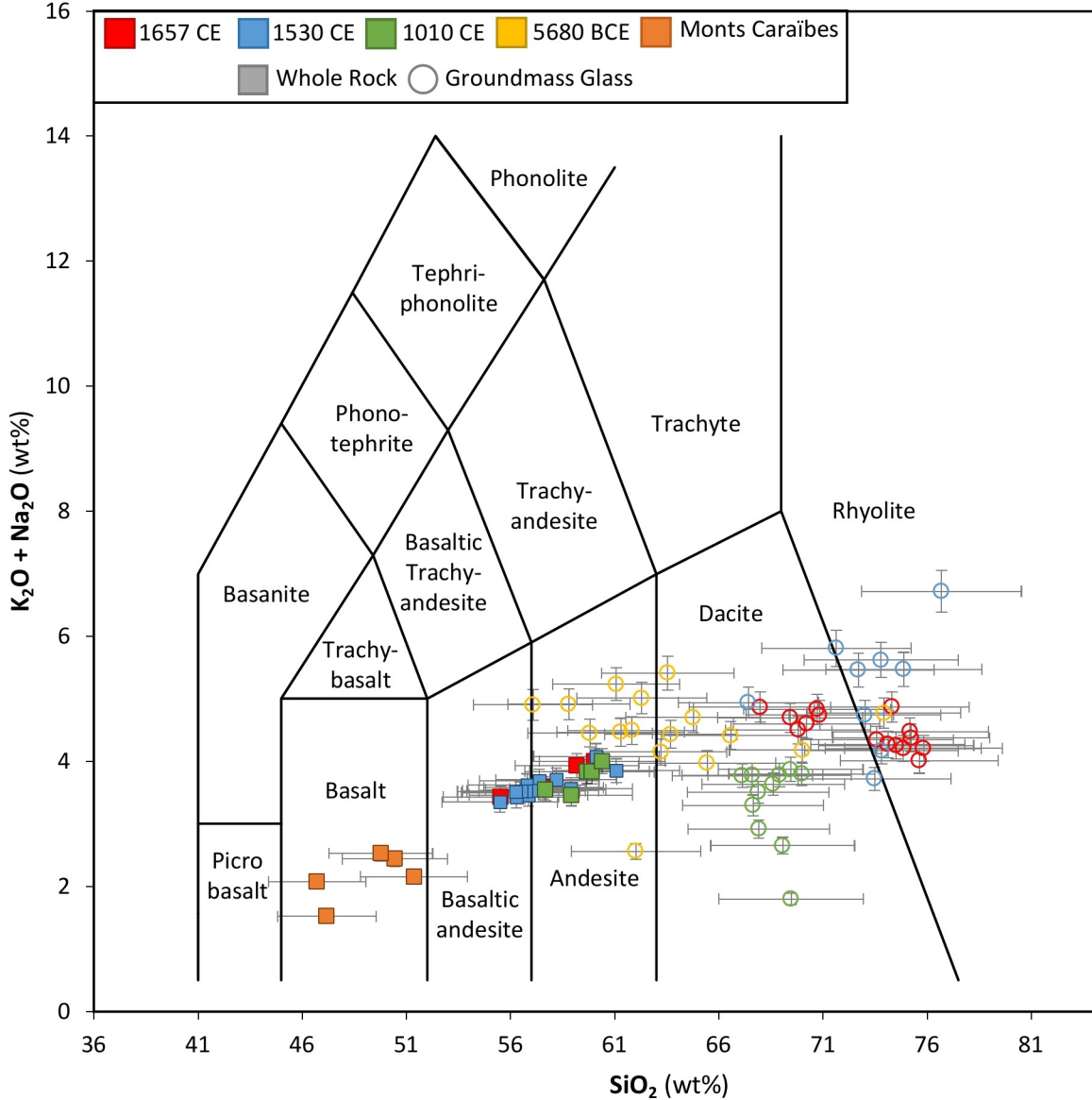
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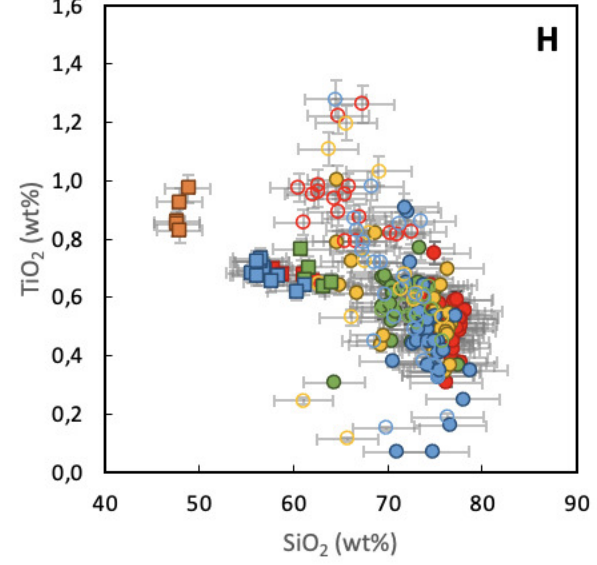
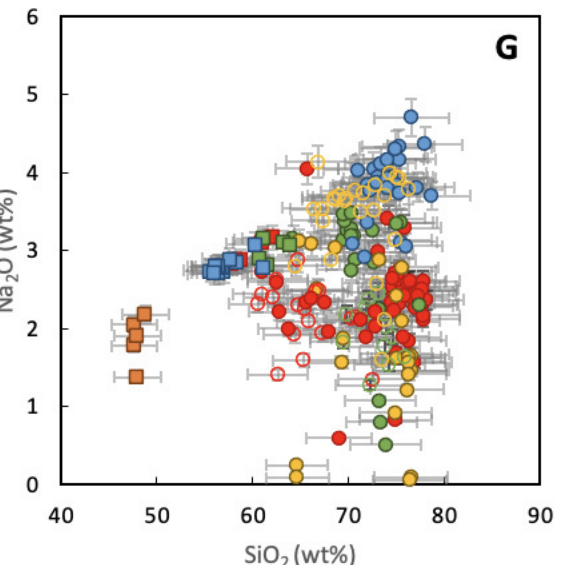
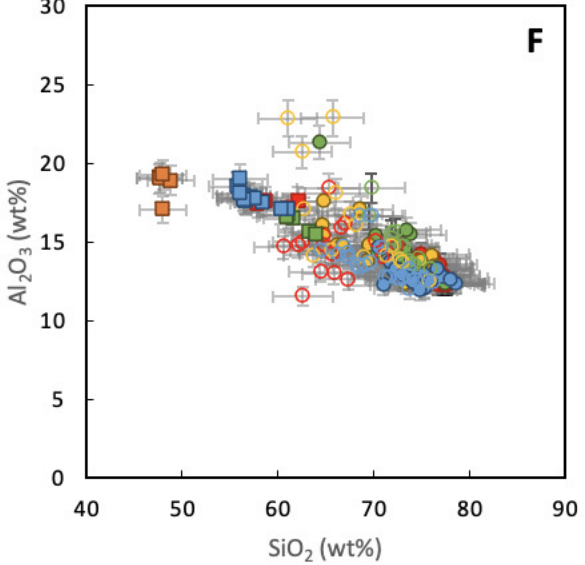
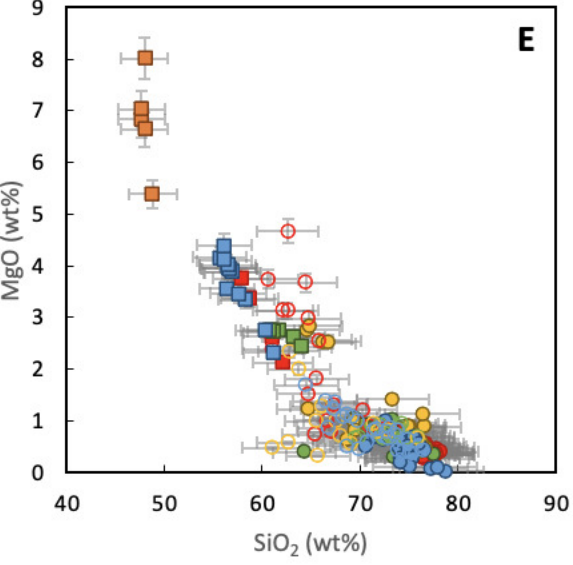
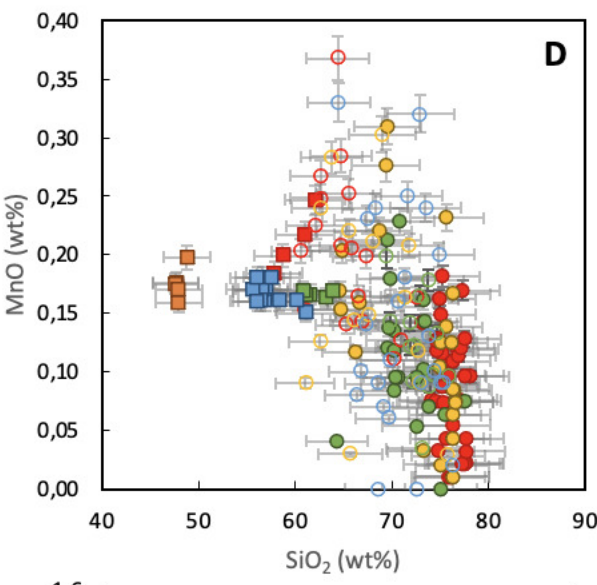
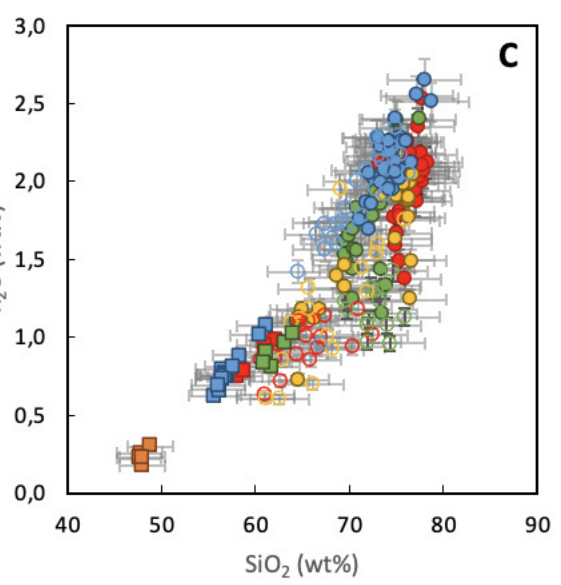
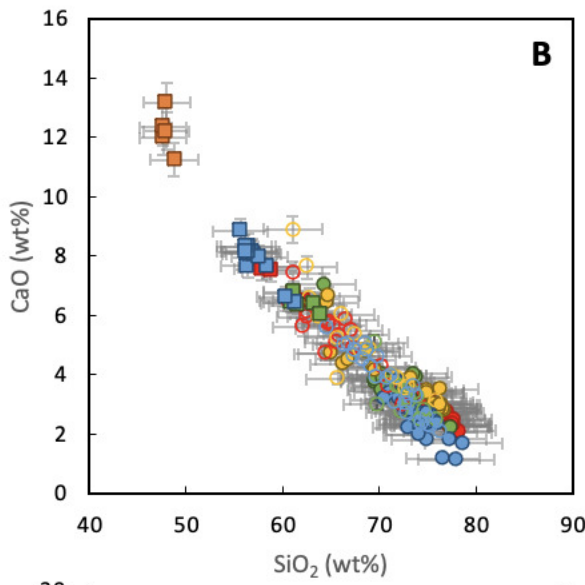
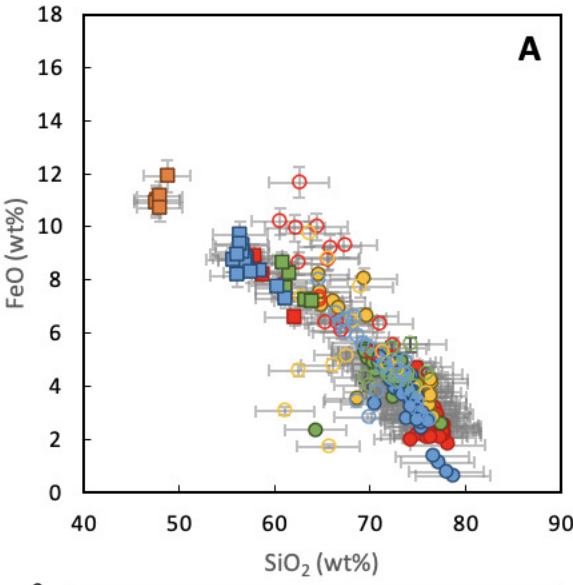
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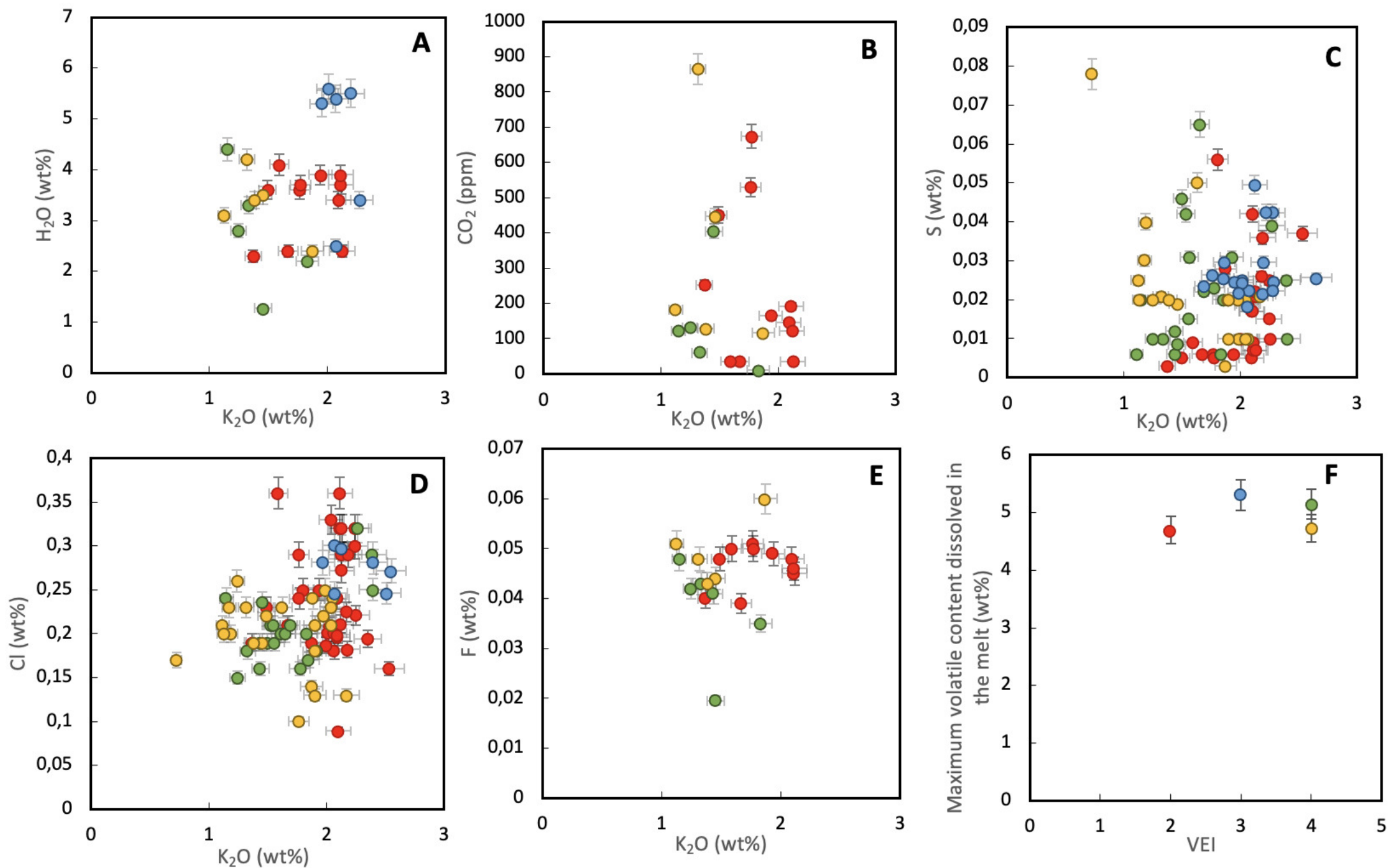








● 1657 CE ● 1530 CE ● 1010 CE ● 5680 BCE



● 1657 CE ● 1530 CE\* ● 1010 CE ● 5680 BCE

